

*ARMY RESEARCH LABORATORY*



**Frequency Control Symposia -  
Program Booklets and Abstracts Predating  
the First Published Proceedings**

**Arthur Ballato**

ARL-TR-1127

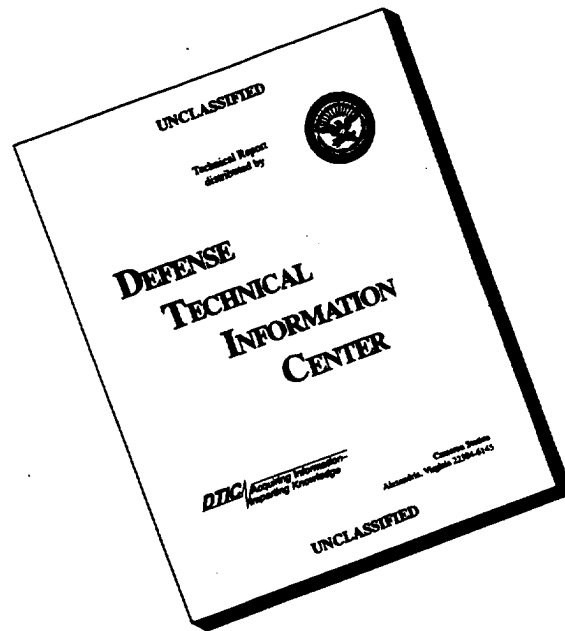
January 1997

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| 13. ABSTRACT (Maximum 200 words)<br><br>The Frequency Control Symposium celebrated its Golden Anniversary in 1996. It began as an annual review of technical progress, largely by those who had been wartime contractors, and was held under the aegis of the Signal Corps Engineering Laboratories at Fort Monmouth, NJ. The first published proceedings started with the tenth. Program booklets have been recovered for five symposia predating the tenth; their contents are published, for historical purposes, in this report.                                                                                                                                                                                      |                                                             |                                                            |                                                                |                                                      |
| 14. SUBJECT TERMS<br><br>Frequency control; resonators; quartz; oscillators;<br>piezoelectrics                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                                            |                                                             |                                                            | 15. NUMBER IF PAGES- IMAGES<br>131                             |                                                      |
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|    |                                                                           |     |
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|----|---------------------------------------------------------------------------|-----|

# FREQUENCY CONTROL SYMPOSIA - PROGRAM BOOKLETS AND ABSTRACTS PREDATING THE FIRST PUBLISHED PROCEEDINGS

## Abstract

The Frequency Control Symposium celebrated its Golden Anniversary in 1996. It began as an annual review of technical progress, largely by those who had been wartime contractors, and was held under the aegis of the Signal Corps Engineering Laboratories at Fort Monmouth, NJ. The first published proceedings started with the tenth. Program booklets have been recovered for five symposia predating the tenth; their contents are published, for historical purposes, in this report.

## Introduction

During its first fifty years, the symposium now officially called the "IEEE International Frequency Control Symposium" has been known by a variety of similar names, each name containing the words "Frequency Control." For our purposes here, we will simply refer to it as the "Frequency Control Symposium," or "FCS." The fiftieth FCS featured an historical session [1-5] to furnish long-term perspective; one saw both breathtaking progress, and at the same time, the continued persistence of various generic technical barriers. Published proceedings began with the tenth (1956); both historical and futuristic papers have appeared over the years, particularly at the 35th [6-11], and again at the 40th symposia [12,13].

Although the FCS began publishing proceedings with the tenth, program booklets are extant for symposia numbers 4, 5, 7, 8, and 9. Those for the 4th and 5th contain only the titles of papers; those for the 7th, 8th, and 9th also contain abstracts. These titles, but particularly the abstracts, provide valuable historical insight into the early developments of our discipline, and are provided in the Appendices for this purpose. Gaps in the paginations of the reproduced booklets denote, in the originals, otherwise blank pages either marked "NOTES," or pages containing the title and author of an unsubmitted abstract; these latter are contained in the program.

In the 4th FCS, held at the Fort Monmouth Officers' Club, we find even then a search for quartz substitutes. Martin Buerger discussed synthesis

of nepheline,  $(\text{Na}, \text{K})\text{AlSiO}_4$ . Walter G. Cady, by then an Emeritus Professor, also gave a talk. (This was apparently his last, until the special session in honor of his 90th birthday at the 1964 FCS. At that time he delivered some short remarks, I recall, to the effect that physics in his day could be carried out by single investigators, whereas today (1964) some of the abstracts from the high-energy physics community had more authors than words.)

The 5th FCS booklet contains notations in the hand of Eduard A. Gerber. Here we find the first hint of quantum frequency control with a paper from Princeton (probably by R. H. Dicke) on the widths of microwave absorption lines.

In the 7th FCS we find papers by Ramond D. Mindlin, Karl S. Van Dyke, Virgil Bottom, Bob Dicke, Clifford Frondel, and Rustum Roy (when Penn State University was still a College).

The 8th FCS contains another Dicke paper, but also papers by Ed Gerber and Art Warner. The abstract of Ed's paper listed research work on, among other things, 'Activity dips in VHF crystal units as influenced by geometry of the blanks, the surface finish and the mounting structure;' these considerations are still pertinent. Arthur W. Warner, Jr. (December 11, 1915 - June 27, 1996), who attended the 50th FCS, reported at the 1954 FCS on reduction of resonator aging by diminishing contamination, use of glass enclosures, polishing of the crystal, and application of 'compact gold electrodes.'

By the 9th FCS, quantum frequency control was beginning to take off, with papers by Fritz Reder, Bob Dicke, and Jerrold Zacharias. Art Warner gave a talk on high precision measurements (one part per billion), with a discussion of noise and phase jitter.

It is hoped that the material contained in the Appendices provides insight into the development of the frequency control profession.

## References

- [1] A. Ballato, "Introduction to the Historical Session," IEEE Intl. Frequency Control Symp. Proc. (50th Annual), Honolulu, HI, June 1996, pp. 4-23.

[2] N. D. Bhaskar, J. White, L. A. Mallette, T. A. McClelland, and J. Hardy, "A Historical Review of Atomic Frequency Standards Used in Space Systems," IEEE Intl. Frequency Control Symp. Proc. (50th Annual), Honolulu, HI, June 1996, pp. 24-32.

[3] M. E. Frerking, "Fifty Years of Progress in Crystal Frequency Standards," IEEE Intl. Frequency Control Symp. Proc. (50th Annual), Honolulu, HI, June 1996, pp. 33-46.

[4] J. R. Norton, J. M. Cloeren, and P. G. Sulzer, "Brief History of the Development of Ultra-Precise Oscillators for Ground and Space Applications," IEEE Intl. Frequency Control Symp. Proc. (50th Annual), Honolulu, HI, June 1996, pp. 47-57.

[5] A. O. McCoubrey, "History of Atomic Frequency Standards; A Trip Through 20th Century Physics," IEEE Intl. Frequency Control Symp. Proc. (50th Annual), Honolulu, HI, June 1996, pp. 1225-1241.

[6] V. E. Bottom, "A History of the Quartz Crystal Industry in the USA," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, pp. 3-12.

[7] A. Ballato, "The Future of the Quartz Crystal Industry - Worldwide," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, pp. 576-582.

[8] J. H. Staudte, "The Future of the Crystal Industry - World Wide," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, pp. 583-591.

[9] W. H. Horton, "Future of the Quartz Industry - World Views," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, p. 592.

[10] T. Takeuchi, "Future of the Quartz Crystal Industry World Views," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, pp. 593-594.

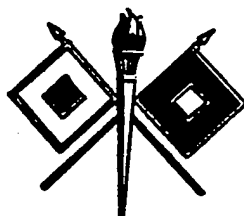
[11] R. Fischer, "Current Trends and Future Projections in the Crystal Industry Worldwide," Proc. 35th Annual Frequency Control Symposium, Philadelphia, PA, May 1981, p. 595.

[12] A. Ballato, "Fortieth Annual Frequency Control Symposium - Award Banquet Remarks," Proc. 40th Annual Frequency Control Symposium, Philadelphia, PA, May 1986, pp. 4-5. See also: A. Ballato, "Frequency and Time Sources - Past, Present, and Future," Japanese Journal of Applied Physics, Vol. 24, (1985) Supplement 24-1, pp. 9-12.

[13] Special Session, "Reminiscences of Early Frequency Control Activities in Honor of the 40th Anniversary of the Frequency Control Symposium," Proc. 40th Annual Frequency Control Symposium, Philadelphia, PA, May 1986, pp. 6-25.

Appendix 1. Symposium Program, Fourth Annual Frequency  
Control Review of Technical Progress

**SYMPOSIUM PROGRAM**  
**FOURTH ANNUAL FREQUENCY CONTROL**  
**REVIEW OF TECHNICAL PROGRESS**  
**26-27 APRIL 1950**



**SIGNAL CORPS ENGINEERING LABORATORIES**  
**FORT MONMOUTH, N.J.**

# SYMPOSIUM PROGRAM

## FOURTH ANNUAL REVIEW OF TECHNICAL PROGRESS

SIGNAL CORPS ENGINEERING LABORATORIES

FREQUENCY CONTROL BRANCH

FORT MONMOUTH, NEW JERSEY

26-27 April 1950

Gibbs Hall, Fort Monmouth Officers' Club

WEDNESDAY, 26 APRIL 1950

*Morning Session* 9:00 a.m. - 12:30 p.m.

Opening Orientation Remarks, Signal Corps  
Engineering Laboratories - E. W. Johnson,  
Chief, Frequency Control Branch, Introducing  
Colonel W. A. Beasley, Deputy Commander,  
Signal Corps Engineering Laboratories

9:00 a.m. - 9:15 a.m.

### *Synthetic Crystal Investigations*

Introduction - H. H. Waesche, Chairman

9:15 a.m. - 9:20 a.m.

1. Synthesis of Tourmaline - Baird Associates-9:20 a.m. - 9:45 a.m.

2. Crystal Synthesis and Twinning Studies  
University of Minnesota

9:45 a.m. - 10:10 a.m.

3. Synthesis of Nepheline  
Edward Washken Laboratories;  
Address by Dr. M. J. Buerger,  
Professor, Mineralogy and Petrography,  
Massachusetts Institute of Technology

10:10 a.m. - 11:00 a.m.

### *Intermission*

4. Quartz Synthesis Studies -  
Antioch College

11:10 a.m. - 11:45 a.m.

5. Synthesis of Quartz and Other  
Crystal Studies - Brush  
Development Company

11:45 a.m. - 12:30 p.m.

Luncheon, Green Room, Gibbs Hall

12:40 p.m. - 1-20 p.m.

## Afternoon Session

1: 30 p.m. - 5: 30 p.m.

6. Summary of Synthetic Quartz  
Investigations - Bell Telephone  
Laboratories

1: 30 p.m. - 1: 45 p.m.

Summary and General Discussion,  
"Synthetics"

1: 45 p.m. - 2: 00 p.m.

## Frequency Control Development

Introduction - W. L. Doxey, Chairman

2: 00 p.m. - 2: 05 p.m.

1. Address: Piezoelectricity as a Branch  
of Thermodynamics - Dr. W. G. Cady,  
Professor Emeritus, Department of  
Physics, Wesleyan University

2: 05 p.m. - 2: 55 p.m.

General Discussion

2: 55 p.m. - 3: 05 p.m.

## Intermission

2. Investigation Overtone Crystal Units  
(50-150 mc) - Radio Corporation of  
America

3: 20 p.m. - 3: 45 p.m.

3. Investigation of Contoured Metal  
Plated Crystal Units (Low Frequency) -  
Radio Corporation of America

3: 45 p.m. - 4: 10 p.m.

4. Development of Thinner Saw Blades -  
The Norton Company

4: 10 p.m. - 4: 25 p.m.

5. Development of Improved Sawing  
Equipment - P. R. Hoffman Company

4: 25 p.m. - 4: 35 p.m.

6. Miniaturized Solder-in Crystal Units  
Bliley Electric Company

4: 35 p.m. - 4: 50 p.m.

7. High Temperature Crystal Units -  
August E. Miller  
500 kc Package Oscillator -  
August E. Miller

4: 50 p.m. - 5: 10 p.m.

General Discussion

5: 10 p.m. - 5: 30 p.m.

## *Evening Program*

Cocktails

6:00 p.m. - 7:00 p.m.

Dinner

7:00 p.m.

### Speakers:

Toastmaster, Lt. Colonel William M. Young,  
Director,  
Squier Signal Laboratory  
introducing:

Major General Francis H. Lanahan,  
Commanding General,  
Fort Monmouth

Dr. Donald H. Menzel,  
Professor of Astrophysics,  
Harvard University,  
"Action on the Sun,"  
with motion picture

Green Room, Gibbs Hall

THURSDAY, 27 APRIL 1950

*Morning Session*

9:00 a.m. - 12:30 p.m.

*Frequency Control Research*

Introduction - W. L. Doxey,  
A. C. Prichard, Chairmen

9:00 a.m. - 9:05 a.m.

1. Detwinning of Crystalline Quartz -  
National Bureau of Standards

9:05 a.m. - 9:25 a.m.

2. Theoretical Studies and Crystal  
Measurements - Wesleyan University

9:25 a.m. - 10:00 a.m.

3. Investigation of Geometric Factors  
Affecting Quartz Crystal Units -  
Tufts College

10:00 a.m. - 10:25 a.m.

*Intermission*

10:25 a.m. - 10:40 a.m.

4. Factors Affecting the Reactance Curve  
of Crystal Units - Colorado Agricultural  
and Mechanical College

10:40 a.m. - 11:15 a.m.

5. Magnetostriction Devices and  
Oscillator Circuits - Armour  
Research Foundation

11:15 a.m. - 11:40 a.m.

6. Stroboscopic X-ray Studies of Oscillating  
Crystals - Pennsylvania State College

11:40 a.m. - 12:10 p.m.

General Discussion

12:10 p.m. - 12:30 p.m.

Luncheon, Green Room, Gibbs Hall

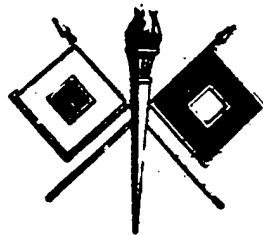
12:40 p.m. - 1:25 p.m.

Appendix 2. Symposium Program, Fifth Annual Frequency  
Control Review of Technical Progress

# **SYMPOSIUM PROGRAM**

**FIFTH ANNUAL FREQUENCY CONTROL  
REVIEW OF TECHNICAL PROGRESS**

**1-3 MAY 1951**



**SIGNAL CORPS ENGINEERING LABORATORIES  
FORT MONMOUTH, N.J.**

# FIFTH ANNUAL REVIEW OF TECHNICAL PROGRESS

## SIGNAL CORPS ENGINEERING LABORATORIES FREQUENCY CONTROL BRANCH FORT MONMOUTH, NEW JERSEY

1 - 3 May 1951

Berkeley-Carteret Hotel, Asbury Park, N.J.

TUESDAY, 1 MAY 1951

Morning Session 11:00 a.m. - 12:15 p.m.

Opening Orientation Remarks, Signal Corps  
Engineering Laboratories - E. W. Johnson,  
Chief, Frequency Control Branch, Introducing  
Lt. Col. Wm. M. Young, Director, Squier  
Laboratory, Introducing Brig. Gen.  
Harry Reichelderfer, Commanding General,  
Signal Corps Engineering Laboratories

11:00 a.m. - 11:15 a.m.

## SYNTHETIC CRYSTAL INVESTIGATIONS

- a* 1. Crystal Synthesis Activities, Signal  
Corps Engineering Laboratories *Stanley - Waseche* 11:15 a.m. - 11:45 a.m.
- a* 2. Growth of Pegmatite Minerals  
Washington University *Frederickson* 11:45 a.m. - 12:15 p.m.
- Inacheon* 12:15 p.m. - 1:30 p.m.
- Afternoon Session 1:30 p.m. - 5:30 p.m.
- a* 3. Crystal Synthesis and Twinning  
Studies, University of Minnesota 1:30 p.m. - 2:00 p.m.
- a* 4. Quartz Synthesis, Antioch College 2:00 p.m. - 2:30 p.m.
- Intermission 2:30 p.m. - 2:45 p.m.
- a* 5. Synthesis of Quartz Crystals Brush  
Development Company *Hale* 2:45 p.m. - 3:30 p.m.
- a* 6. Synthesis of Quartz at High Temperatures  
and Pressures, Bell Telephone Laboratories *Walker* 3:30 p.m. - 4:15 p.m.
7. Question and Answer Session on Signal  
Corps Report Requirements. *Doxey* 4:15 p.m. - 4:30 p.m.
- General Discussion 4:30 p.m. - 5:30 p.m.

## FREQUENCY CONTROL STUDIES

- |   |     |                                                                                                     |                         |
|---|-----|-----------------------------------------------------------------------------------------------------|-------------------------|
| c | 8.  | Effect of Crystal Geometry on the Slope of the Reactance Curve, Colorado A&M                        | 9:00 a.m. - 9:30 a.m.   |
| b | 9.  | Theoretical Investigation of Partially Plated Crystals, Wesleyan University                         | 9:30 a.m. - 10:00 a.m.  |
| b | 10. | Detwinning of Quartz<br>National Bureau of Standards                                                | 10:00 a.m. - 10:15 a.m. |
|   |     | Intermission                                                                                        | 10:15 a.m. - 10:30 a.m. |
| b | 11. | Crystal Studies Utilizing Stroboscopic X-Rays, Penn. State College                                  | 10:30 a.m. - 11:00 a.m. |
| e | 12. | Effect of Plating on Aging Characteristics of Crystals, Georgia Institute of Technology             | 11:00 a.m. - 11:30 a.m. |
| g | 13. | Theoretical Investigation of Oscillator Circuits, University of Illinois                            | 11:30 a.m. - 11:45 a.m. |
| b | 14. | Precision Electro-mechanical Filters, Tafts College                                                 | 11:45 a.m. - 12:15 p.m. |
|   |     | Luncheon                                                                                            | 12:15 p.m. - 1:30 p.m.  |
| g | 15. | Very High Frequency Oscillator Circuits, Georgia Institute of Technology                            | 1:30 p.m. - 2:15 p.m.   |
| c | 16. | Improvement of Heat Dissipation Characteristics of Crystal Units, Colorado A&M                      | 2:15 p.m. - 2:45 p.m.   |
|   |     | Intermission                                                                                        | 2:45 p.m. - 3:15 p.m.   |
| b | 17. | Reduction of the Doppler Contribution to Widths of Microwave Absorption Lines, Princeton University | 3:15 p.m. - 3:30 p.m.   |
| b | 18. | Use of Scintillating Crystals for the Detection of Soft X-rays, University of Oregon                | 3:30 p.m. - 4:00 p.m.   |
| b | 19. | Relations Between Current, Voltage, and Amplitude of Vibration in Piezoelectric Crystals            | 4:00 p.m. - 4:30 p.m.   |
|   |     | Discussion                                                                                          | 4:30 p.m. - 5:30 p.m.   |

*Evening Program*

Dinner

7:00 p.m.

**Speakers:**

Toastmaster, Lt. Colonel William M. Young

**Principal Speaker:**

Dr. Frederick H. Pough  
American Museum of  
Natural History  
"The Glamorous Crystal"  
with slides.

Crystal Terrace

Berkeley Carteret Hotel

THURSDAY, 8 MAY 1951

Morning Session 9:00 a.m. - 12:30 p.m.

MANUFACTURING AND TESTING

- g 21. 500 KC Packaged Oscillator as  
Developed by A. E. Miller 9:00 a.m. - 9:15 a.m.
- j 22. Contouring Equipment as Developed  
by Bausch & Lomb Company 9:15 a.m. - 9:30 a.m.
- j 23. Precision Quartz Saw as Developed  
by P. R. Hoffman Company 9:30 a.m. - 9:45 a.m.
- j 24. Thin Saw Blades as Developed by  
the Norton Company 9:45 a.m. - 10:00 a.m.
- Intermission 10:00 a.m. - 10:15 a.m.
- d 25. Practical Application of  
Proportional Temperature  
Control Devices 10:15 a.m. - 10:30 a.m.
- i 26. Use of the Crystal Impedance  
Meter in the Laboratory and  
in Production 10:30 a.m. - 10:45 a.m.
- n 27. Consideration of Drive Levels  
in the CR-18, CR-23 and Other  
Crystal Units 10:45 a.m. - 11:15 a.m.
- i 28. Apparatus and Method for  
Production Testing of Plated  
Type Crystal Units Hunt  
Corporation 11:15 a.m. - 12:30 p.m.

Afternoon Session 2:00 p.m. - 5:00 p.m.

Luncheon 12:30 p.m. - 2:00 p.m.

- n 29. General Discussion of MIL-C Type  
Specifications Including a Dis-  
cussion of CR-4/U Type Crystals  
Bell Telephone Laboratories 2:00 p.m. - 2:30 p.m.
- n 30. Low Frequency Crystals in HC-6/U  
Holders, Radio Corporation of  
America 2:30 p.m. - 2:45 p.m.
31. General Discussion of  
Manufacturing Problems 2:45 p.m. - 5:30 p.m.

Appendix 3. Symposium Program, Seventh Annual Frequency  
Control Review of Technical Progress

—symposium—program—

*7th Annual*

FREQUENCY CONTROL REVIEW  
of

TECHNICAL PROGRESS

18 - 20 MAY 1953

**SIGNAL CORPS ENGINEERING LABORATORIES**

**FORT MONMOUTH, N. J.**

SYMPOSIUM PROGRAM  
SEVENTH ANNUAL REVIEW OF TECHNICAL PROGRESS  
SIGNAL CORPS ENGINEERING LABORATORIES  
FREQUENCY CONTROL BRANCH - CHEMICAL PHYSICS BRANCH  
FORT MONMOUTH, NEW JERSEY

18, 19, 20 May 1953

Berkeley-Carteret Hotel, Asbury Park, N. J.

*Outline of Meetings*

*Monday, 18 May 1953*

|            |                         |                 |
|------------|-------------------------|-----------------|
| 9:00 a.m.  | Registration            | Palm Terrace    |
| 11:00 a.m. | General Session         | Crystal Terrace |
| 12:15 p.m. | Luncheon                |                 |
| 1:30 p.m.  | Piezoelectric Vibrators | Crystal Terrace |

*Tuesday, 19 May 1953*

|            |                                                |                 |
|------------|------------------------------------------------|-----------------|
| 8:30 a.m.  | Frequency Control Devices and Applications     | Crystal Terrace |
| 12:15 p.m. | Luncheon                                       |                 |
| 1:30 p.m.  | Fundamental Properties of Crystals             | Crystal Terrace |
| 3:00 p.m.  | Research and Development in the United Kingdom | Crystal Terrace |
| 3:45 p.m.  | Quartz Synthesis                               | Crystal Terrace |
| 7:00 p.m.  | Annual Dinner                                  | Crystal Terrace |

*Wednesday, 20 May 1953*

|            |                                                   |                 |
|------------|---------------------------------------------------|-----------------|
| 8:30 a.m.  | Circuitry and Test Equipment                      | Crystal Terrace |
| 9:00 a.m.  | Crystal Chemistry and Growing Techniques          | Solarium        |
| 10:45 a.m. | Crystal Unit Production Forum                     | Crystal Terrace |
| 12:15 p.m. | Luncheon                                          |                 |
| 1:30 p.m.  | Crystal Unit Production Forum (Contd.)            | Crystal Terrace |
| 1:30 p.m.  | Crystal Chemistry and Growing Techniques (Contd.) | Solarium        |

## Detailed Schedules

MONDAY MORNING, 18 May 1953

### General Session

- 11:00 a.m.      Introductory Program      Signal Corps Engineering Laboratories  
Mr. W. L. Doxey, Chief, Frequency Control Branch,  
Squier Signal Laboratory  
Lt. Col. Robert K. Saxe, Director, Squier Signal Laboratory  
Brig. Gen. Edwin R. Petzing, Commanding, Signal Corps  
Engineering Laboratories  
Dr. Harold A. Zahl, Director of Research, Signal Corps  
Engineering Laboratories
- 12:15 p.m.      Luncheon

MONDAY AFTERNOON, 18 May 1953

### Piezoelectric Vibrators

- 1. 1:30 p.m.      Investigations in the Mathematical Theory of Vibrations of  
Anisotropic Bodies - R. D. Mindlin - Columbia University
2. 2:00 p.m.      Piezoelectric Crystal Studies - K. S. Van Dyke - Wesleyan  
University
- 3. 2:30 p.m.      Research Investigations on Fundamental and Overtone Crystals  
V. E. Bottom - Colorado A&M College
- 4. 3:00 p.m.      Ring Resonators - L. G. Chase - Motorola, Inc.  
3:30 p.m.      Intermission
- 5. 3:45 p.m.      Aging Study of Metal Plating on Quartz Crystals - R. Belser  
Georgia Institute of Technology
- 6. 4:15 p.m.      Improvement in Crystal Units for Precise Frequency Control -  
R. A. Sykes - A. W. Warner - J. P. Griffin - Bell Telephone  
Laboratories
- 7. 4:45 p.m.      Drive Level, Sputtering, and X-Ray Studies - E. A. Gerber,  
R. Morris, S. S. Brody - Frequency Control Branch, SCEL,  
Fort Monmouth, N. J.

TUESDAY MORNING, 19 May 1953

### Frequency Control Devices and Applications

1. 8:30 a.m.      Low Frequency Electromechanical Filters - S. P. Lapin -  
Motorola, Inc.
2. 9:00 a.m.      Characteristics of Electromechanical Filters - C. R. Mingins  
A. D. Frost - Tufts College

- 3. 9:30 a.m. *g* Transistor Circuit Applications to Frequency Control -  
E. Gonzalez - Frequency Control Branch, Squier Signal  
Laboratory
- 4. 10:00 a.m. *g* Frequency Control Above 150 mc/sec - D. Fraser - Georgia  
Institute of Technology
- 10:30 a.m. Intermission
- 5. 10:45 a.m. *g* Frequency Control Systems - R. W. Frank - R. W. Stuart -  
General Radio Co.
- 6. 11:15 a.m. *f* Frequency and Time Interval Standards - W. D. George -  
National Bureau of Standards
- 7. 11:45 a.m. *f* Molecular Absorption Phenomena - R. H. Dicke - Princeton  
University - Radio Corporation of America
- 12:15 p.m. Luncheon

TUESDAY AFTERNOON, 19 May 1953

*Fundamental Properties of Crystals*

- 1. 1:30 p.m. *b* Surface Structure of Quartz Crystals - G. W. Arnold -  
Naval Research Laboratories
- 2. 2:00 p.m. *b* X-Ray Diffraction Studies of Piezoelectric Crystals -  
R. Pepinsky - The Pennsylvania State College
- 3. 2:30 p.m. *b* Radiation and Physical Properties of Crystals - C. Frondel -  
Harvard University

*Research and Development in the United Kingdom*

- 4. 3:00 p.m. Review of Research and Development in the United Kingdom -  
H. T. Mitchell - British Post Office
- 3:30 p.m. Intermission

*Quartz Synthesis*

- 5. 3:45 p.m. *a* Status of Raw Quartz - H. H. Waesche - Frequency Control  
Branch, Squier Signal Laboratory
- 6. 4:00 p.m. *a* Growth of Quartz Crystals at High Pressures - G. T. Kohman -  
A. C. Walker - Bell Telephone Laboratories
- 7. 4:30 p.m. *a* Growth of Quartz at Low Pressures - D. R. Hale -  
W. H. Charbonnet - Brush Laboratories Co.
- 8. 5:00 p.m. *a* Twinning in Synthetic Quartz - J. W. Gruner - University of  
Minnesota

TUESDAY EVENING, 19 May 1953

7:00 p.m. Annual Dinner, Crystal Terrace, Berkeley Carteret Hotel  
Guest Speaker - Dr. I. M. Levitt, Director Fels Planetarium  
Franklin Institute, Phila., Pa.  
Subject: "Space Travel - The Human Being in  
Space"

WEDNESDAY MORNING, 20 May 1953

*Circuitry and Test Equipment*

1. 8:30 a.m. *i* Crystal Impedance Meter TS-710/TSM - R. Green - Radio Frequency Laboratories
2. 8:50 a.m. *i* A Direct Reading Frequency Meter - P. G. Hansel - Servo Corporation of America
3. 9:15 a.m. *j* Crystal Oscillators in the VHF and UHF Regions - G. I. Davies - Davies Laboratories - Naval Research Laboratories
4. 9:40 a.m. *j* Theory of Oscillator Circuits - J. W. Hoffman - B. Silverman - C. V. Jakowatz - University of Illinois
5. 10:00 a.m. *j* Oscillator Design Considerations for Military Equipment - E. A. Roberts - Armour Research Foundation - Wright Air Development Center
- 10:30 a.m. Intermission

*Crystal Unit Production Forum*

6. 10:45 a.m. Procurement - Philadelphia Signal Corps Supply Agency
7. 11:15 a.m. Inspection - Philadelphia Signal Corps Supply Agency
8. 11:45 a.m. Specifications - Bureau of Ships
- 12:15 p.m. Luncheon

WEDNESDAY AFTERNOON, 20 May 1953

1. 1:30 p.m. Crystal Requirements for Communications Equipment - Collins Radio Company
2. 2:00 p.m. *c* Pressure-mounted VHF Crystals - Bliley Electric Company
3. 2:30 p.m. Training Aids and Production Short Cuts - Hunt Corporation
- 3:00 p.m. Intermission
4. 3:15 p.m. *i* The Human Factor and Crystal Testing - Pioneer Electric & Research Corp.
5. 3:45 p.m. *j* Crystal Production Problems Associated with Air Force Equipment Design - Wright Air Development Center
6. 4:00 p.m. General Discussion

WEDNESDAY MORNING, 20 May 1953

*Crystal Chemistry and Growing Techniques*

*Joint Session - Frequency Control Branch - Chemical Physics Branch*

1. 9:00 a.m. *a* Studies of Silica Transfer and Diffusion - R. G. Yalman - Antioch College
2. 9:20 a.m. *a* Silica Structures - M. J. Buerger - Edward Washken Laboratories
3. 9:40 a.m. *a* Composition of Fluid Inclusions in Minerals - E. Roedder - University of Utah
4. 10:00 a.m. *a* Pegmetite Studies - A. F. Frederickson - J. E. Cox - Washington University
- 10:20 a.m. Intermission
5. 10:30 a.m. *a* Stability Relations in Barium Titanate and Other Minerals - R. Roy - E. F. Osborn - The Pennsylvania State College
6. 11:00 a.m. *a* Growth of Barium Titanate - H. C. Kremers - The Harshaw Chemical Company
7. 11:30 a.m. *a* Ferromagnetic Perovskite Compounds - D. Heinz - Polytechnic Institute of Brooklyn
- 12:15 p.m. Luncheon

WEDNESDAY AFTERNOON, 20 May 1953

1. 1:30 p.m. *a* Polymorphism in Natural Micas - E. W. Heinrich - A. A. Levinson University of Michigan
2. 1:50 p.m. *a* Synthetic Mica Progress - R. A. Hatch - J. E. Comeforo - U. S. Bureau of Mines
3. 2:10 p.m. *b* Infrared Studies of Crystals - G. B. B. M. Sutherland - University of Michigan
- 2:40 p.m. Intermission
4. 2:55 p.m. *b* Infrared Spectroscopy of Crystals - R. S. Halford - Columbia University
5. 3:25 p.m. *b* Magnetochemistry of Crystals - P. W. Selwood - Northwestern University
6. 3:45 p.m. *a* Preparation of Ultra High Purity Germanium - W. E. Metcalf - R. K. Riel - C. E. Smith - Eagle-Pitcher Company
7. 4:05 p.m. *a* High Purity Silicon - F. B. Litton H. C. Anderson - Foote Mineral Company

Appendix 4. Abstracts, Seventh Annual Frequency  
Control Review of Technical Progress

## SURFACE STRUCTURE OF QUARTZ CRYSTALS

G. W. ARNOLD

NAVAL RESEARCH LABORATORIES

Measurements of the thickness of distorted surface layers on crystals have been made. Those crystals studied were prepared with abrasives ranging from F carborundum to aluminum and iron oxides. Electron diffraction photographs were made of the ground surfaces and the change in these patterns observed as the distorted layers were etched away. The criterion for determination of the depth of distortion was taken as that depth at which Kukuchi line patterns begin to form. Etching curves were established by means of a very sensitive balance and the thickness of material removed computed from these data. Some comparison between natural and synthetic quartz with respect to changes in surface structure by abrasive action and etching are given.

## AGING STUDY OF METAL PLATING ON QUARTZ CRYSTALS

R. B. BELSER

GEORGIA INSTITUTE OF TECHNOLOGY

Regardless of whether deposited by evaporation, sputtering or electroplating, thin metal films appear to be subjected to states of strain and disorder not so predominant in a metal cooling in bulk from a molten condition. A susceptibility to change, therefore, is inherent in the nature of the growth of the films. Driven by surface and thermodynamic forces, and corrosion, changes occur specific to a metal film and its environment. Upon these changes frequency shifts of plated quartz resonators are dependent.

From the standpoint of the stability of the coated crystals, sputtered gold films have given superior results. Evaporated gold, after artificial aging, has behaved comparably. Evaporated aluminum, naturally and artificially aged, has given excellent stability. Evaporated and sputtered silver films, although especially susceptible to corrosion and aggregation, have given good stability and high performance when properly handled. Limited tests of electroplated silver and nickel films indicate reasonable stability.

Of the other metals deposited, copper, indium and palladium have been found inferior whereas sputtered platinum gave excellent stability. Dual layer metal films, subject to diffusion and alloying at the metal interface, gave poor stabilities. Silver, overcoated with nickel, appears to be a stable combination and leads may be attached with tin-lead solder and a flux.

The dynamic resistance parameters of crystals coated by evaporation are superior to those sputtered with accordingly higher ultimate expected "Q" values. Values above 150,000 for 6 mc crystals are not unusual, whereas ultimate values above 400,000 appear possible by use of the coating and aging procedures now recognized as superior.

V. E. BOTTOM

COLORADO A&amp;M COLLEGE

A large number of measurements of the L and C of harmonic crystal units have been made. The measurements were made on crystal units similar to type CR-23 at frequencies from thirty to ninety megacycles per second, and from the third to the ninth harmonic. The accuracy of the measurements is believed to be approximately 5%. As predicted by theory, the values of L are nearly independent of h the harmonic order, but experimental values are larger than predicted values probably because of the uncertainty in the size of the effective vibrating area of the quartz plate. A remarkable degree of uniformity is obtained among units fabricated to the same geometric specifications.

Temperature frequency curves on CR-23 type units made from synthetic quartz (AT cut) are unlike those obtained from units made from natural quartz. The principal differences are: (1) the ZZ' angles must be increased for synthetic quartz, (2) the turning points occur at higher temperatures in synthetic quartz, (3) the frequency decreases more rapidly at low temperatures in synthetic quartz, and (4) all units show a marked decrease in activity at low temperatures. At room and high temperatures, the activity is similar to that obtained from natural quartz.

The ZZ' angle for optimum performance of CR-23 type units is approximately 35°26' with natural quartz. Plates made from one sample of quartz from Brush had to be cut at 35°31', and those from a sample of Bell quartz at 35°36'.

It appears that synthetic quartz offers possibilities for high temperature operation. Units have been fabricated from both Brush and Bell quartz having frequency deviations of less than 10 ppm(.001%) in the temperature range -25°C to +110°C.

## SILICA STRUCTURES

M. J. BUEGER

EDWARD WASHKEN LABORATORIES

A *derivative structure* is one derived from a simpler *basic structure* by generalization. This can occur either by distortion of the simpler structure, or by substitutions for its atoms by others of different chemical species. When the substitute atoms have smaller valences than those for which they substitute, this must be compensated by addition of other atoms which can be described as stuffing atoms. Only such atoms as can be accommodated in the available voids of the structure are acceptable stuffing atoms.

This paper treats of the stuffed derivatives of quartz, tridymite, and cristobalite.

Quartz has voids of limited sizes. These are too small for K or Na, but large enough for Li, Be, or B. Eucryptite,  $\text{LiAlSiO}_4$ , and  $\text{LiAlSi}_2\text{O}_6$  are known derivatives of quartz.

Both tridymite and cristobalite have voids large enough to house Na, K,

or Ca. The best known derivatives of tridymite are nepheline,  $\text{NaK}_3\text{Al}_4\text{Si}_4\text{O}_{16}$ , and the several forms of  $\text{KAlSiO}_4$ . The compound  $\text{K}_2\text{MgSi}_3\text{O}_8$ , and possibly  $\text{K}_2\text{Mg}_5\text{Si}_{12}\text{O}_{10}$ , appear to be derivatives of tridymite in which Mg substitutes for Si.

The cristobalite derivatives include carnegieite,  $\text{NaAlSiO}_4$ , and  $\text{Na}_2\text{CaSiO}_4$ . It seems likely that  $\text{Na}_2\text{MgSiO}_4$  and  $\text{Na}_2\text{BeSiO}_4$  may also assume tridymite-like structures.

Non-silicate derivatives of silica are briefly discussed.

## RING RESONATORS

L. G. CHASE

MOTOROLAR INCORPORATED

Quartz flexural vibrators consisting of rings into which a gap is cut have advantages over the ordinary straight bars. The frequency of vibration of both is proportional to the width and inversely proportional to the mean length squared. For a given piece of quartz, the mean length of a ring can be made to exceed that of the maximum bar by approximately the factor of  $\pi$ . Thus lower frequency vibrators can be obtained from the smaller pieces of natural quartz. Frequency-temperature characteristics are controlled by adjustment of the ring dimensions.

Z slabs of quartz form the base from which the resonators are cut, and flexure is made to occur in the XY plane. Special techniques are required for electrode placement and interconnection as well as mechanical suspension.

This paper discusses the fabrication of the thin Z cut ring, methods of mounting and results of conducted experimentation.

## CRYSTAL OSCILLATORS IN THE VHF AND UHF REGIONS

G. L. DAVIES

THE DAVIES LABORATORIES INC.

Recent developments in the investigation of basic types of VHF overtone crystal oscillators are discussed; the discussion covers design objectives, adjustment and data recording procedure, and typical data for the various circuits under test. These include the cathode coupled, crystal impedance meter, Lister anti-resonant, transformer coupled, bridged-T, and other oscillators.

Techniques are described which are employed in the determination of crystal power dissipation and crystal electrical characteristics (admittance) in the frequency range 20 to 135 mc. Difficulties experienced, especially at the higher frequencies, are touched on, with some mention of the modified techniques required to surmount them.

A VHF-UHF experimental transmitter was recently built for use in demonstrating oscillator circuitry in operating equipment. This transmitter is composed of an overtone oscillator operating in the range of 75 to 135 mc, a

buffer amplifier, a power amplifier, and a power tripler furnishing 15 watts of output in the frequency range 225 to 405 mc. The oscillator-buffer stages are constructed as a compact plug-in assembly and may be readily replaced by other assemblies as they are developed.

## MOLECULAR ABSORPTION PHENOMENA

R. H. DICKE

PRINCETON UNIVERSITY

Reported is a project at Princeton University to attempt to produce a high-Q resonance at 1420 mc in atomic hydrogen and a project at the R.C.A. Laboratories at Princeton to produce a high-Q resonance in ammonia at 24,000 mc. The work at Princeton University is under the supervision of the speaker; the work at R.C.A. is being carried out by Dr. L. E. Norton and Mr. G. W. Leck, with the speaker acting as a consultant.

The Princeton work makes use of two novel principles. First, the atomic hydrogen is in an inert atmosphere which may be helium or, more simply, molecular hydrogen. The collisions of the atomic hydrogen with the inert gas molecules has an effect of strongly reducing the doppler effect contribution to the frequency breadth of the resonance. It is essential that these same collisions do not themselves disturb the precession of the electron spin which is the source of this resonance. The second novel feature is that this experiment does not employ absorption, with the attendant difficulties in balancing out a carrier, but rather makes use of a new principle in microwave spectroscopy, the production of and detection of "super-radiant states" of the gas.

The R.C.A. project is a continuation of the work begun at Princeton on ammonia resonance in a gas cell employing plane parallel wire grids to modulate the molecular resonance frequencies. There have been two important modifications. First, the grid spacing has been doubled, leading to a different mode of operation. Secondly, the C. W. Method has been replaced by a pulsed method employing "super-radiant states."

## FREQUENCY CONTROL SYSTEMS

R. W. FRANK - F. W. STUART

GENERAL RADIO COMPANY

In February, 1952, two systems were proposed by the General Radio Company for the generation of at least 1,000 stable frequency channels of 10 kc width between 100 kc and 10 mc. Such systems will permit transmitter frequency control without the use of a quartz crystal for each channel.

Both of the systems proposed use only one quartz crystal. This crystal is used to derive standard frequencies which are used to phase-lock oscillators. The crystal and a locked oscillator are about the only circuits which are common to the two systems.

The first system which will be discussed has been named the counter system. In this system an oscillator of variable frequency is phase-locked

by the output of a decade-settable counter whose output frequency is compared to a standard frequency. Since the V.F.O. is connected to the decade-settable counter and the comparison frequency is fixed, the V.F.O. frequency is pre-settable in decades. The equation  $f_o/A = f_c$  expresses the principle, where  $f_o$  is the V.F.O. frequency,  $A$  is the count number, and  $f_c$  is the fixed comparison frequency.

In the second system, one oscillator in each decade is phase-locked to one of a group of standard frequencies. The phase lock for the oscillator of each decade is performed at a frequency offset which permits the insertion of the standard frequency derived by the locked oscillators of all lower decades. In this fashion a decade-switching system permits an unambiguous selection of a single output frequency with an accuracy determined only by the number of decades.

## FREQUENCY CONTROL ABOVE 150 MC PER SECOND

D. W. FRASER

GEORGIA INSTITUTE OF TECHNOLOGY

During the past year emphasis has been placed upon two separate and basically distinct processes, or methods, by which precise frequency control may be possible. The first of these methods involves the purely electrical processes of high  $Q$  cavity resonators, the second is associated with the resonances of various nuclei.

Precise frequency control through the medium of a cavity resonator may be obtained only if the cavity utilized exhibits a high  $Q$  and also excellent temperature stability. Coaxial cavities have been constructed which utilize a center conductor formed of silver plating over a glass tubing of low coefficient of expansion. This tubing, whose trade name is Vycor, has a coefficient of expansion of about 0.8 parts per million per degree centigrade. An oscillator which used this cavity as a frequency controlling device exhibited a stability closely comparable to the theoretical stability stated above. To further improve the stability, temperature compensating devices have been added to other cavities, and at present cavities are under construction which will utilize a form of ceramic (Stupalith) with a claimed zero coefficient of expansion.

Nuclear quadrupole resonances were investigated for their suitability as frequency controlling phenomena. An unsuccessful attempt was made to construct an oscillator locked on the response of the sample which was in one arm of a bridge balanced at all frequencies except that of the quadrupole resonance. Several regenerative detectors were constructed which detected quadrupole resonances in halogen compounds in the frequency range from 25 Mc. to 200 Mc. Reasonably large signal-to-noise ratios were observed, but at low signal levels. All of the resonances suffered from a temperature coefficient of approximately one part in ten thousand per centigrade degree. Measurements of the parameters of the responses were attempted with the sample in a passive circuit. Because of the weakness of the responses these experiments yielded but little useful information. Frequency control by quadrupole resonances is apparently not feasible.

Measurement of characteristics of the cavities has presented numerous problems. To facilitate these measurements a Cavity Q Meter has been constructed. This device utilize a frequency swept oscillator, a variable frequency oscillator, and a band-pass amplifier. Their combined use permits a simultaneous CRT presentation of the cavity frequency response curve and of pairs of markers to measure the band width of the response curve. This equipment shows considerable promise in facilitating future measurements of cavity resonators.

## PEGMATITE STUDIES

A. F. FREDERICKSON AND JOSEPH E. COX

WASHINGTON UNIVERSITY

The factors controlling the addition of material onto a crystal, the coagulation of colloidal particles or the cementation of various solids by silicate cements are:

- (a) the electric charge on the particles or molecules,
- (b) their degree of solvation and
- (c) the structural configuration of the surfaces involved.

It is postulated that large quartz crystals grow by the cementation of mosaic units by silicate gels according to a particular mechanism. The role of the positive ions is to electrically neutralize the surface of the particles allowing a close approach which is followed by oxygen-bond formation in place of the previous OH bonds (dehydration).

Mosaics are postulated instead of the sole use of various silicate ions or micelles because of the difficulty of forming regular crystals by suitable cross-linkage polymerizations.

Reactions of this kind are also believed to play an important role in the growth of layer-lattice silicates.

## RADIATION AND PHYSICAL PROPERTIES OF CRYSTALS

C. FRONDEL

HARVARD UNIVERSITY

The primary concerns of our contract work on quartz has been (1) the identification of the mechanism by which the chemical composition of quartz varies, and (2) a correlation of the chemical variation with variation in the unit cell dimensions, indices of refraction, density and irradiation-response. A selected group of five natural quartz specimens has been examined by high-precision methods in the above regards, and restricted measurements have been made on a series of synthetic quartz samples prepared under controlled conditions.

An important aspect of our recent work has been the development of new and improved instruments for the precision measurement of the indices of refraction and of the unit cell dimensions. An instrument based on the Emmons

double-variation technique has been developed for the measurement of the indices of crushed grains to a precision of about 0.00002, comparable to the precision obtained by the conventional minimum deviation method on large single-crystal prisms. A new and simple x-ray powder diffraction camera has been developed that yields a precision of better than 1 part in 55,000 under optimum conditions. Precision unit cell measurements have been made of NaCl and ThO<sub>2</sub>, as test substances, and new data on quartz, earlier measured to lower precision, are being obtained.

A correlation and final interpretation of our data on quartz has not yet been effected, primarily because of the incomplete state of our work on the density and unit cell dimensions. It can be stated, however, that there is a significant variation in the chemical composition of quartz, primarily of the nature of a coupled solid solution of alkalies and aluminum for silicon, and that the observed variation in the density, indices of refraction and unit cell dimensions is dependent thereon.

## FREQUENCY AND TIME INTERVAL STANDARDS

W. D. GEORGE

NATIONAL BUREAU OF STANDARDS

A brief description will be given of the more recent improved techniques, components and systems incorporated in the National Standard of Frequency, e.g. a more uniform basic reference of time interval and quartz crystal units having greater constancy. Performance of the present standard will be discussed.

Activities at the National Bureau of Standards which are expected to result in better frequency standards (basic and working type) will be mentioned, e.g., improved temperature control, operation of crystal units at lower temperatures and transistor oscillators.

## DRIVE LEVEL, SPUTTERING, AND X-RAY STUDIES

7, 53

E. A. GERBER, R. MORRIS, S. S. BRODY

FREQUENCY CONTROL BRANCH, SCEL, FONT MONMOUTH

An outline of the internal research and development program on frequency control components is presented.

It is shown that, by applying proper cleaning methods during the plating operation, the influence of power dissipation upon crystal resistance can be minimized. Frequency perturbations due to increasing power level can be avoided by the use of heat distributing electrodes.

A method and equipment are demonstrated to increase or decrease the frequency of a plated crystal at choice. Decrease is accomplished by the known sputtering method, increase by reversing the voltage and ion-bombarding the crystal.

Crystals with different abrasive finishes were progressively etched in ammonium bifluoride, and rocking curves were taken after each stage of etch

to determine the order of the lattice misorientation. Polished crystals presented a minimum of misorientation after 3 minutes of etching corresponding to an etch rate of  $0.03F^2$ , semipolished after 13 minutes ( $0.25F^2$ ), and 3000 mesh crystals after 17 minutes ( $0.35F^2$ ). Optical transmission, and the equivalent resistance of 1000 mesh, 3000 mesh, semi-polished and polished crystals as a function of etch are also discussed.

## TRANSISTOR CIRCUIT APPLICATIONS TO FREQUENCY CONTROL

E. GONZALEZ

FREQUENCY CONTROL BRANCH, SCCL, FORT MONMOUTH, N. J.

The physical nature and basic circuit properties of transistors are summarized, leading to the discussion of several circuit applications currently under study. The use of transistors as switching elements in counters, multi-vibrators and similar circuits is considered, including the advantages and present limitations. Transistor amplifier and oscillator circuits are discussed and an experimental frequency meter is demonstrated which indicates the urgent logistic need for semi-conductor devices meeting stringent military requirements. The present status and trends of transistor developments are evaluated.

## CRYSTAL IMPEDANCE METER TS-710( )/TSM

R. GREEN

## RADIO FREQUENCY LABORATORIES

This Crystal Impedance Meter is designed to measure the equivalent circuit parameters of piezoelectric crystals in the frequency range of 10 to 1100 kilocycles. The paper will describe the development and operational uses of the Crystal Impedance Meter TS-710( )/TSM.

The development began with a circuit that the Bell Telephone Laboratories had used for checking crystal units in this range. With certain revisions and additions the unit was JANized and RF voltmeter and ohmmeter circuits were added. The Crystal Impedance Meter will measure the resistance of crystal units under either series- or anti-resonant conditions, and, by use of the built-in voltmeter, the power dissipated in the crystal unit may be computed very easily.

This Crystal Impedance Meter will check crystal units (within its frequency range) mounted in HC-6/U and HC-13/U holders. It will also check crystal units with octal bases, using the adapter that is supplied. There are two other adapters included; a 500,000 and a 50,000 ohm variable resistor, along with 14 fixed calibrating resistors.

# TWINNING IN SYNTHETIC QUARTZ

J. W. GRUNER

UNIVERSITY OF MINNESOTA

Since our last symposium much time has been spent in an attempt to correlate electrical twinning and optical twinning, which also is recognizable as "fibrous" twinning. The fibres are recognized by their interference colors and, as their directions correspond to those ordinarily associated with optical twinning (as will be shown in photomicrographs) they have been designated as such. Electrical twinning in crystals grown at the University of Minnesota is nearly all microscopic in size. As it is "interfingered" and coexistent with the optical and "fibrous" twinning, it is difficult to detect unless the etched surfaces are very carefully prepared. The twinned individuals frequently start at or near the seed and the new orientation will gradually expand and "crowd" out the other, resulting in what appears like an almost untwinned crystal on an etched surface a few millimeters above the seed. In this respect, synthetic quartz grown at the University of Minnesota is different from that of Brush Laboratories Co. which has been examined as a part of this investigation. It is possible that the smaller bombs introduce important changes in the growth behavior.

## GROWTH OF QUARTZ CRYSTALS AT LOW PRESSURES

7, 53

D. R. HALE, W. H. CHARBONNET, J. M. JOST

BRUSH LABORATORIES COMPANY

During the last 12 months the facilities for growing an increased quantity of synthetic crystals was completed and put in full operation. A total of about 210 lbs. of synthetic quartz was produced prior to March 1953. The mechanical operation of the equipment gave no trouble. The temperature control over long periods still needs improvement. The standard process required 2M  $\text{Na}_2\text{CO}_3$ , a growing temperature of about  $350^\circ\text{C}$  and 5000 lbs. per sq. in. pressure. A 50 lb. batch of crystals requires a growing period of about two months. The visual quality is generally good except for a layer at the junction of seed and new growth. However, a certain amount of optical twinning has occurred. In this research part of the program, pressures down to 1500 psi have been successful, yielding about half the standard growth rate or 0.3 mm/day measured normal to the minor rhombohedral face. In view of this success, two autoclaves welded from double extra heavy steel pipes are under construction. High quality quartz has been grown in sodium hydroxide solution; however, the rate is much less than in sodium carbonate.

In the development of a process for growing radio-grade quartz, it became apparent that the conditions under which crystals can be grown at an acceptable rate may be best within a relatively small range of temperature, pressure, and concentration of alkali, but that actually quartz can be deposited under a very wide range of conditions. Solubility studies were begun to broaden our knowledge of these matters.

Two molar  $\text{Na}_2\text{CO}_3$  was used at temperatures up to  $450^\circ\text{C}$  and pressures up to 10,000 psi. The method involves combining alkaline solution and quartz in a steel vessel, heating at a predetermined temperature sufficiently long to

attain equilibrium, quenching, and analyzing the solutions. It was confirmed that solubility increases with both temperature and pressure, and that over much of the range two phases, one of high and one of low density, are formed. The system is ternary ( $\text{Na}_2\text{CO}_3\text{--H}_2\text{O--SiO}_2$ ) below about  $290^\circ\text{C}$ , in the region where one liquid phase is present, and quaternary ( $\text{Na}_2\text{O--CO}_2\text{--H}_2\text{O--SiO}_2$ ) in the region where two liquid phases are present. A three-dimensional model has been constructed to represent this system of five variables. A few experiments were made on mixtures of  $\text{Na}_2\text{CO}_3$  and  $\text{NaOH}$ .

## INFRARED SPECTROSCOPY OF CRYSTALS

R. S. HALFORD

COLUMBIA UNIVERSITY

(Unavailable)

## DIRECT READING FREQUENCY METER

P. G. HANSEL

SERVO CORPORATION OF AMERICA

Frequency is measured by determining the position of an unknown frequency along a frequency scale. The frequency scale is a spectrum of crystal controlled harmonics of fine and coarse spacing.

The operation is fully automatic. A probe scans the coarse spectrum between a known starting frequency, or "milepost", and the unknown frequency. As the probe passes each spectrum component, a count pulse is delivered to the appropriate decade of a mechanical counter. When the unknown is reached, the probe reverses and counts the number of fine-spectrum components between the unknown and the last coarse spectrum component. A "sliding harmonic" interpolator may be used for greater measuring resolution.

All functions, except counting, are performed electronically. Compact telephone stepping relays capable of 100 counts per second are used for counting. A novel circuit has recently been devised for combining the functions of spectrum generation probing and coincidence detection.

The frequency range is 100 kc to 100 mc, the accuracy 0.001% and the time required for measurement 3 to 4 seconds. The frequency meter has 28 tubes, a volume of 1.5 cubic feet and weighs 35 pounds.

## SYNTHETIC MICA PROGRESS

R. A. HATCH AND J. E. COMEFORO

U. S. BUREAU OF MINES

The Army Signal Corps - Bureau of Mines cooperative synthetic mica program has had for its primary aim during the current fiscal year (1) the investigation of methods of growing crystals and for consolidating reconstituted synthetic mica sheets, and (2) the fabrication and study of

various types of mica ceramics. Crystallization experiments with the internal electric-resistance heating furnace have resulted in some improvement in the quality but not in the size of the mica sheets produced. To date, the most promising solution to the sheet mica problem is a reconstituted-heat treated mixture of two synthetic micas having different melting points.

Synthetic mica ceramics, a by-product of the original research objectives, are now on the threshold of commercial production. These new materials, which are soft, machinable, and mechanically strong, may be fabricated by many different techniques. In addition to their established merit as a new ceramic dielectric and as a substitute for block talc, recent work indicates their applicability as a high temperature inorganic bonding agent and structural material. Synthetic mica cermets may well become one of the most important products developed as a result of this program.

### POLYMORPHISM IN NATURAL MICAS

E. WM. HEINRICH AND ALFRED A. LEVINSON

UNIVERSITY OF MICHIGAN

Micas belong mainly to the monoclinic system; some are hexagonal; a few triclinic types have been reported. Most muscovites possess the 2-layer monoclinic structure, but some phengites (high-Si muscovites) have formed as the 3-layer hexagonal polymorph. Muscovite may accommodate as much as about 3.3%  $\text{Li}_2\text{O}$  without noticeable structural deformation; lithian muscovite (as much 3.7%  $\text{Li}_2\text{O}$ ) shows slight deviation from a 2-layer monoclinic form. With increasing amounts of  $\text{Li}_2\text{O}$ , mixed-layer structures appear between lithian muscovite and a 6-layer monoclinic lepidolite. The 6-layer type alone forms for  $\text{Li}_2\text{O} = 4.0 - 5.1\%$ ; above  $\text{Li}_2\text{O} = 5.1\%$  lepidolite appears as the 1-layer monoclinic form. Thus in this series polymorphism is a function of Li content, for increasing numbers of Li atoms occupying normally vacant positions in the muscovite structure increase the perfection of the structure to permit alternative stackings. A 3-layer, hexagonal, uniaxial lepidolite polymorph is formed by twinning of the 1-layer monoclinic type. Roscoelite has a 1-layer monoclinic structure which is apparently concomitant with the V-Al substitution. Phlogopites are monoclinic, with 1- and 2- layer types, or hexagonal with a 3-layer stacking; the 1-layer polymorph is most common. Biotites also show 1- and 2-layer monoclinic forms, a 3-layer hexagonal form, and 6-, 24- and 48-layer triclinic types have been reported. In the phlogopite-biotite series there is no apparent relationship between composition and polymorphism, nor any obvious relationship between temperature of formation and structure. However pegmatitic biotites from a single district may be principally of one structure type, indicating perhaps some environmental control.

### FERROMAGNETIC PEROVSKITE COMPOUNDS

D. HEINZ

POLYTECHNIC INSTITUTE OF BROOKLYN

Two series of mixed oxides of lanthanum and the transition elements are being studied in an attempt to obtain ferromagnetic materials with perovskite structures similar to those obtained by Jonker and van Santen.

The first of these series consists of compounds of the following type  $\text{La}(\text{M}^{\text{III}})_{1-x}(\text{N}^{\text{III}})_x\text{O}_3$ , in which  $\text{M}^{\text{III}}$  and  $\text{N}^{\text{III}}$  refer to two different transition metal ions in valence state (III). This series is illustrated by the group of compounds  $\text{La Mn}_{1-x} \text{Cr}_x \text{O}_3$  which have been studied in some detail.

The second series contains compounds of the type  $\text{La}_{1-x} \text{A}_x^{\text{II}} \text{M}_{1-x}^{\text{III}} \text{M}_x^{\text{IV}} \text{O}_3$ , where M refers to a transition element and A refers to strontium or barium. The formula given above is idealized. These are in reality defect structures. An example of this type of compound is  $\text{La}_{1-x} \text{Sr}_x \text{Fe}_{1-x}^{\text{III}} \text{Fe}_x^{\text{IV}} \text{O}_3$ .

All compounds were prepared by solid state reactions at temperatures varying from  $900^\circ \text{C}$  to  $1200^\circ \text{C}$ . Qualitative tests for ferromagnetism were made with a small Alnico magnet and some of these compounds have been found to be weakly ferromagnetic. X-ray powder photographs were studied for variation with composition.

## THEORY OF OSCILLATOR CIRCUITS

J. W. HOFFMAN, B. SILVERMAN, C. V. JAKOWATZ

UNIVERSITY OF ILLINOIS

A method is demonstrated for predicting the action of a triode oscillator whose output is almost sinusoidal. By this method the fundamental amplitude and frequency, as well as the harmonic content, of the output may be determined. The general procedure involves the assumption of a harmonic solution. The expression for the triode characteristic is experimentally determined and the differential equation of the circuit is reduced to a set of algebraic equations by the substitution of the assumed harmonic solution. The various frequency components are separated, giving rise to a set of recurrence relations for the harmonic coefficients. Substitution of these recurrence relations in the circuit equations yields a complex equation which may be separated and solved for the fundamental frequency and amplitude. The method is applied to a tuned-plate oscillator, and design curves are obtained. Also, a procedure is outlined which permits the results to be corrected for the effects of current flow in the grid circuit. Tables are given for the comparison of predicted and experimental results, and good agreement is demonstrated. A discussion is included indicating the applicability of the method to a large class of oscillators.

## GROWTH OF QUARTZ CRYSTALS AT HIGH PRESSURES

7,53

G. T. KOHMAN - A. C. WALKER

BELL TELEPHONE LABORATORIES

Effort at the Bell Telephone Laboratories has been directed towards growth of quartz crystals at relatively high pressures and temperatures. It has been found that solubility of quartz in alkaline solution at a given temperature increases with pressure, thus establishing favorable conditions for crystal growth. Pressure is self generated by expansion of the solution, reaching approximately 12 to 15 thousand psi at  $400^\circ \text{C}$ . During the past year

the problems of spurious seeding, crystal inclusions, corrosion and autoclave geometry have received attention. Spurious seeding has been found to vary considerably with the composition of the solvent. Crystal inclusions and the join between seed and new growth appear to be related to gas content and corrosion. Corrosion difficulties have been minimized by means of improved metal plating techniques. The possibility that they are related to the gaseous oxygen content of the autoclave is under investigation. An improved type of welded closure has reduced leakage difficulties. The process appears capable of producing crystals of high quality weighing a pound or more in approximately two months time.

## GROWTH OF BARIUM TITANATE

H. C. KREMERS

HARSHAW CHEMICAL COMPANY

This paper is a report on the current status of barium titanate growth as presented at the recent conference held at Squier Signal Corps Laboratories. The Harshaw Laboratories have, for several years, studied the growth of macro crystals of barium titanate grown in massive boule form by use of melts substantially in stoichiometric proportions. By slight variations of composition, control of tetragonal end product was assured. During the past year M.I.T. has produced some boules almost colorless. This was in part achieved by the use of very pure materials. Phase studies carried out at Penn. State College have generally verified the belief that crystals produced from pure melts in substantially stoichiometric proportions are grown in the hexagonal form and then go through a solid hexagonal-cubic phase change upon passing through the 1500-1400°C range. Some crystals grown in the Harshaw Laboratory have retained the hexagonal form at room temperature when melts were kept deficient in  $\text{TiO}_2$ . The hexagonal-cubic phase change, unless carried out under almost ideal conditions, produces tetragonal crystals at room temperatures, containing multiple domains. Of the several hundred boules grown in the Harshaw Laboratories, only one or two indicated promise as regards multiple domain components. These experiments were "accidents" and could not be duplicated. In the light of these facts, it appears that more hope of success can be attained by growing barium titanate macro crystals from various mixtures so designed as to keep the growth temperature well below the hexagonal-cubic phase transition. Mixtures of this type have usually consisted of  $\text{BaCl}_2$  flux with  $\text{BaCO}_3$  and  $\text{TiO}_2$  added. A working formula much used has been:

50 gms.  $\text{BaCl}_2$   
25 gms.  $\text{BaCO}_3$   
6 gms.  $\text{TiO}_2$

Such a mixture is usually heated to 1200-1250°C and allowed to cool slowly. Crystals of barium titanate are rapidly formed in thin plates, usually under 1 mm cross section. Currently, variations of the above formula are being tried. Substitution of  $\text{BaBr}_2$  in place of  $\text{BaCl}_2$  has resulted in materially increasing crystal size.

## LOW FREQUENCY ELECTROMECHANICAL FILTERS

S. P. LAPIN

MOTOROLA, INCORPORATED

Electro-mechanical filters, composed of mechanically vibrating members in conjunction with electrical to mechanical converting elements, and in certain instances using electrical components, have several advantages over corresponding conventional inductance-capacitance filters. These advantages include a selectivity curve with a flatter acceptance nose and having steeper rejection slopes. Such a filter would also be a relatively small and light unit, would have increased stability, and possibly could be sealed and permanently adjusted at the factory. Mechanical resonators, in the frequency range from 100 KC to 1 MC, can be used as the resonant elements in a filter structure, with suitable mechanical coupling between them. Suitable converting elements at both ends of the structure are required. Resonators, either piezoelectric or magnetostrictive, can also be used in conjunction with coils and condensers. The resonators are described in terms of their equivalent electrical parameters, and this equivalent circuit can then be incorporated into an electrical filter structure. The theoretical and experimental data presented covers both types of these filters.

## HIGH PURITY SILICON

F. B. LITTON - H. C. ANDERSON

FOOTE MINERAL COMPANY

The preparation of high purity silicon through the thermal decomposition of silicon tetraiodide was investigated in both a static and intermittent flow system. The metal produced by the standard vanArkel-deBoer technique showed that its purity was a function of impurities in the source material and is not entirely satisfactory for electronic usage. When high purity Electro-Metallurgical silicon was used as the source material, drawn single crystals ranged in resistivity from 0.5 to 3.0 ohm-cm. By using iodide refined material for the silicon source, drawn single crystals ranged in resistivity from 3.0 to 8 ohm-cm. Silicon of somewhat higher purity was prepared by iodinating Electro-Metallurgical silicon, fractionally distilling the tetraiodide in a packed quartz column, and subsequently decomposing the purified tetraiodide by vaporizing it over a heated tantalum wire under high vacuum in a intermittent flow system. In the distillation operation, elements such as boron appear to be segregated in the first distillate, while aluminum and heavy elements appear to be segregated in the still pot. Melted single crystal silicon produced from tetraiodide distilled once through a 3.7 plate column, at 200 mm. pressure, has a resistivity of 18 ohm-cm., whereas doubly distilled tetraiodide had resistivity of 30 ohm-cm. Work on the fractional distillation of silicon tetraiodide and its thermal decomposition is continuing. Equipment is described for producing metal in 50-gram quantities.

# PREPARATION OF ULTRA HIGH PURITY GERMANIUM

W. E. MEDCALF - R. K. RIEL - C. E. SMITH

THE EAGLE-PICHER COMPANY

Germanium metal was prepared by reduction of germanium (IV) chloride with (a) zinc vapor and (b) hydrogen. Zinc vapor proved to be a more efficient reductant, but higher purity germanium was produced using hydrogen as a reductant. Reducing  $\text{GeCl}_4$  with zinc vapor, using a modification of the method of Rick and McKinley <sup>(1)</sup>, yielded 85 per cent of theoretical. The germanium was obtained as needles and contained zinc as the chief impurity. The zinc contamination was reduced below spectrographic detection by acid leaching followed by vacuum melting. Progressive crystallization of the molten germanium segregated the zinc impurity and produced metal averaging 20 ohm-centimeters resistivity and having none of the P type characteristics which might be expected if zinc were present. Germanium metal of higher purity was prepared by the reduction of  $\text{GeCl}_4$  with hydrogen. The products of this reaction were a function of the reactor temperature and the partial pressure of hydrogen. Metal recovery ranged from 2 to 40 per cent of theoretical. The only impurity detectable spectrographically in the metal was silicon - about 2 parts per million - and occasionally a trace of magnesium. This represented greater purity than was found in the oxide prepared by hydrolysis of the tetrachloride. The physical and electrical properties of the germanium produced by hydrogen reduction of  $\text{GeCl}_4$  were functions of the reactor temperature. Above  $950^\circ\text{C}$ ., ingots of metal were formed with resistivities normally between 5 and 10 ohm-cm, but ranging as high as 30 ohm-cm. Below  $930^\circ\text{C}$ ., germanium coatings were formed. Coatings ranging from 10 to 75 microns in thickness and having desirable electrical properties were deposited on graphite, lavite and quartz. The coatings deposited below  $800^\circ\text{C}$ . were P type, while coatings deposited between 800 and  $930^\circ\text{C}$ . were N type. Heat treatment of the germanium coatings on graphite markedly affected the current-voltage characteristics.

## INVESTIGATIONS IN THE MATHEMATICAL THEORY OF VIBRATIONS OF ANISOTROPIC BODIES

7, 53

R. D. MINDLIN

COLUMBIA UNIVERSITY

The purpose of this contract was to support an effort to advance the mathematical theory of the vibrations of crystals. During the year in which the contract was in effect, four advances were made in the theory of thickness-shear and flexural vibrations of plates:

- (1) Equations of motion of crystal plates of varying thicknesses were developed. Solutions were obtained for plates having the cross-section of a double wedge and for plates with beveled edges.
- (2) The influence of electrode size on the frequency spectrum was investigated. A problem was solved which shows the effect of coupling between coated and uncoated portions of a crystal plate.

(1) C. E. Rick and T. D. McKinley - G.S.R.D. Contract No. OEMsr 1139

- 175
- (3) The problem of suppression of overtones was studied. A solution was obtained which gives the electrode shape which excites only one mode of cylindrical motion in a rectangular plate.
  - (4) Equations of motion in polar coordinates were deduced and the problem of vibrations of an isotropic circular disk was solved. The computation of the frequencies is not yet completed.

## CHARACTERISTICS OF ELECTROMECHANICAL FILTERS

C. R. MINGINS - A. D. FROST

TUFTS COLLEGE

An investigation has been made of the variations in the parameters of the effective network of a single-crystal piezoelectric filter at frequencies in the one- to ten-megacycle range as physical changes in the figure of the plate are introduced. The behavior of the motional inductances, capacitances, and resistances can be considered in terms of the mechanical effects of inertia and elasticity, but the electromechanical effect must also be taken into account and is often the preponderant influence. The appropriate manipulation of the parameters in this way results in a considerable control of the bandwidth. In the use of a single-crystal filter for wider bands, however, difficulties in matching impedances can produce an objectionable sag in the transmission characteristic within the pass band between the fundamental thickness shear and the particular higher mode chosen for the upper boundary<sup>1</sup>. The introduction of additional resonances cannot be a remedy, as is shown by an analysis of the effective network. Such an analysis indicates that pass regions can exist only between pairs of resonances, and these regions are set off by zeros from the remainder of the transfer function. Although this limitation also extends to such arrangements as the combination of filter units in parallel or the addition of lumped-impedance elements, nevertheless units can be combined by special methods so as to improve the characteristic by "nesting" additional peaks in the depressed region.

## REVIEW OF RESEARCH AND DEVELOPMENT IN THE UNITED KINGDOM

H. T. MITCHELL

BRITISH POST OFFICE

(Unavailable)

<sup>1</sup> C. R. Mingins, S. Bartnoff, and L. A. Howard, Phys. Rev. 88, 154 (1952)

# X-RAY DIFFRACTION STUDIES OF PIEZOELECTRIC CRYSTALS

R. PEPINSKY

PENNSYLVANIA STATE COLLEGE

The advantages of combined X-ray and neutron single-crystal diffraction measurements, and the limitations of both methods, are illustrated in discussions of the structural mechanisms of the ferroelectric transitions in  $\text{KH}_2\text{PO}_4$ , Rochelle salt and other tartrates, and  $\text{BaTiO}_3$ , and non-ferroelectric transitions in  $\text{NH}_4\text{H}_2\text{PO}_4$  and  $\text{ND}_4\text{D}_2\text{AsO}_4$ . Fourier maps are shown of  $\text{KH}_2\text{PO}_4$ , obtained both from X-ray and neutron data. The strong role of hydrogens in the Rochelle salt transitions is established by means of a comparison of X-ray and neutron scattering observations. The extension of these methods to an accurate analysis of quartz is discussed.

## OSCILLATOR DESIGN CONSIDERATIONS FOR MILITARY EQUIPMENT

E. A. ROBERTS

ARMOUR RESEARCH FOUNDATION

The feasibility of standardizing crystal oscillator design is under investigation. Studies to date indicate that performance data can be presented in a form which may lead to the development of a standard method of cataloging the operational characteristics of crystal oscillator circuits. Examples of such performance data, similar in form and intent to published vacuum tube characteristics, are presented. These data include typical performance and frequency stability curves of two anti-resonant circuits, utilizing MIL-C-3098 type crystals, operating in the 2 to 16 mc range. The analysis, measurement, and construction of the curves are explained, and applications of the curves for circuit comparison and performance prediction will be considered. Techniques for sealing crystal units free from moisture have been investigated. Experiments have been conducted that relate crystal resistance and frequency change to the amount of moisture sealed in an HC-6 unit. From these measurements and substantiating calculations the maximum allowable moisture has been determined. The effect of variations in crystal heating rate during the dewpoint test has been studied. Several crystal unit sealing methods are evaluated.

## COMPOSITION OF FLUID INCLUSIONS IN MINERALS

E. ROEDDER

UNIVERSITY OF UTAH

Practically every synthetic or natural quartz crystal other than the best optical grade contains some fluid filled inclusions, and most natural white quartz contains on the order of  $(10)^9$  such inclusions, averaging about 2 microns in diameter, in every cc. In some cases these inclusions can be proven to be a true sample of the original medium from which deposition of the quartz crystal took place, trapped during growth of the crystal; in others they are evidently samples of fluid present in the quartz during the healing of fractures formed after the original crystallization. In either event,

these inclusions preserve for us actual samples of fluids from which quartz has grown in nature. A technique has been developed for the extraction and limited wet chemical and spectrographic analysis of the salts in solution in the fluids of these inclusions. The possibilities and limitations of the technique are discussed, and results given for quartz samples from a number of types of occurrence.

## STABILITY RELATIONS IN BARIUM TITANATE AND OTHER MINERALS

R. ROY - E. F. OSBORN

PENNSYLVANIA STATE COLLEGE

During the year considerable progress has been recorded in the obtaining and application of phase equilibrium data to the problems of the growth of crystalline phases. Complete phase equilibrium data are presented for the systems  $\text{BaO-TiO}_2$ ,  $\text{CaO-TiO}_2$ ,  $\text{TiO}_2\text{-SiO}_2$ , and  $\text{CaO-TiO}_2\text{-SiO}_2$ . Due to the importance of the polymorphism of  $\text{BaTiO}_3$  in its growth from melts, a number of other pertinent systems have also been investigated. Work in the following systems is in an advanced state and the results will be described:  $\text{BaTiO}_3\text{-SiO}_2$ ,  $\text{BaTiO}_3\text{-NaNbO}_3$ ,  $\text{BaTiO}_3\text{-BaCl}_2$ ,  $\text{BaTiO}_3\text{-CaTiO}_3$  and  $\text{SrO-TiO}_2$ . An extensive investigation into the possibilities and limitations of multiple ionic substitution in the perovskite lattice has been completed and resulted in the discovery of several new compounds. The hydrothermal work has progressed along two main directions. First, data have been obtained in the system  $\text{BaO-TiO}_2\text{-H}_2\text{O}$  to provide a background for the possible hydrothermal growth of these crystals. Second, a survey of a large number of aluminosilicate systems was undertaken to find the existence, stability and ease of formation of micas of these compositions. Finally, due to the applicability of these techniques to hydrate systems, the work on the stability of the third group oxide hydrates was continued. Markedly revised data and new equilibrium diagrams are presented for the systems  $\text{Cr}_2\text{O}_3\text{-H}_2\text{O}$ ,  $\text{In}_2\text{O}_3\text{-H}_2\text{O}$ ,  $\text{Sc}_2\text{O}_3\text{-H}_2\text{O}$ ; new compounds have been found, and older suggested compounds eliminated.

## MAGNETOCHEMISTRY OF CRYSTALS

P. W. SELWOOD

NORTHWESTERN UNIVERSITY

Work has continued on the use of magnetic susceptibility methods to investigate structural relationships in paramagnetic and antiferromagnetic systems. Pure titanium sesquioxide has been shown to be antiferromagnetic, with a Curie point near the specific heat anomaly at  $240^\circ\text{K}$ . Solid solutions of titanium sesquioxide in alumina become progressively more paramagnetic with increasing magnetic dilution. The magnetic moment of the titanium tripositive ion at high magnetic dilution approaches the theoretical value for one unpaired electron, but some phase ratios and compositions show evidence of ferromagnetisms as recently reported by Weise. These results aid in the interpretation of earlier data on the magnetic susceptibility of the titanium-oxygen system at phase ratios between  $\text{Ti}_2\text{O}_3$  and  $\text{TiO}_2$ . Analysis of magnetic data in this and related systems has shown that in

magnetically concentrated systems there is a negative interaction, different from antiferromagnetism, and leading to a diminished effective magnetic moment. There also appears to be a type of positive interaction, or group action, differing from ferromagnetism but leading to enhanced effective magnetic moments. These phenomena are being studied with reference to the hydrolysis of ferric perchlorate, and to similar systems with simultaneously controlled pH and complex formation. The results so far obtained help to explain the anomalous effective magnetic moments found in isopolynuclear complexes of iron and chromium. It is expected that this work will ultimately result in a clearer picture of exchange interaction, and of structure, in crystals, glasses, and amorphous solids.

## INFRA-RED STUDIES OF CRYSTALS

G. B. B. M. SUTHERLAND

UNIVERSITY OF MICHIGAN

Infrared absorption spectra of crystals arise from vibrations of the atoms in the crystalline lattice which are associated with a corresponding periodic change in dipole moment. The vibration spectrum of any crystal lattice cannot yet be predicted theoretically but a good deal of progress has been made in certain special cases. The extent to which the infrared spectra of diamond, silicon and germanium can be interpreted in terms of current theories will be briefly reviewed, with special reference to the problem of the two types of diamond. The explanation of the ferroelectric properties of barium titanate in terms of the structure of the crystal is still uncertain. One theory (Jaynes) predicts an infrared absorption band at  $10\mu$ . No band is present in the spectrum at that wavelength. In micas the positions of the hydrogens have not been determined. It may be possible to do so by infrared techniques. This problem will be discussed. Of considerable interest, also, is the use of infrared as a means of estimating OH groups in various micas.

## IMPROVEMENTS IN CRYSTAL UNITS FOR PRECISE FREQUENCY CONTROL

R. A. SYKES - A. W. WARNER - J. P. GRIFFIN

BELL TELEPHONE LABORATORIES

Condensed results reported by the National Bureau of Standards on several experimental 100 kc GT-cut quartz crystal units, built at the Bell Telephone Laboratories for use as precision standards, are presented. The frequency stability is 0.5 to 5 pp  $10^9$  per day and the Q is  $1.5 \times 10^6$  to  $4 \times 10^6$ . This represents a 10-fold improvement in stability and a 20-fold increase in Q over the performance normally found in commercial crystal units of this type. These improvements result from refinements in constructional detail and advanced processing techniques. A new 5 megacycle primary frequency standard crystal unit, having a Q between 2 and  $3 \times 10^6$ , and an impedance of 100 ohms at resonance, has been developed using an overtone mode of vibration of a specially shaped AT-cut quartz plate. The average frequency change, as shown by data on a number of units from the National Bureau of Standards and from the Bell Telephone Laboratories, is between 0.5 and 3 parts in  $10^9$  per day.

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This crystal unit is comparatively free from the effects of mechanical and thermal shocks. This, together with the short time necessary for stabilization, make it suitable for use as a portable frequency standard.

## PIEZOELECTRIC CRYSTAL STUDIES

K. S. VAN DYKE

WESLEYAN UNIVERSITY

Brief reports are given on several phases of the contract work and on other work in piezoelectricity in this laboratory. Under the first are included: (1) B. H. Camp's critical interpretation of Mindlin's development of the theory of the thickness-shear quartz resonator; (2) the new 3rd edition of the author's *Manual of Piezoelectric Data*; (3) a bibliography of piezoelectricity for 1952; (4) present status of the experimental studies of the distribution of the vibration in thickness-shear quartz resonators; (5) a first attempt to measure Q for a quartz plate which has been relieved of its support and thus is in free fall; (6) the rewriting of the matrix of piezoelectro-dielectric properties in a form judged to be the most convenient as a starting point for the picturing of the elementary dipoles in the crystal. Under the second are described (7) some basic elementary experiments in piezoelectricity for undergraduate students in physics; (8) commentary on two recent lengthy manuscripts by the author as introductions to the field of piezoelectricity, one for a technical audience and one having some slant toward the more popular.

## STATUS OF RAW QUARTZ

H. H. WAESCHE

FREQUENCY CONTROL BRANCH, SCCL, FORT MONMOUTH

Crystalline quartz continues to be the most important material for frequency control in military service because of its unique combination of physical and electrical properties. No equivalent substitute is apparent. For these reasons, the Signal Corps is committed to the use of quartz and, therefore, has taken steps to overcome the shortcomings of supply by searching for other natural sources and by developing means of synthesizing radio grade crystals; in addition, quartz has been stock-piled as a regular part of the National Stockpile Program. Through a combination of these three approaches, a future quartz crystal supply is assured. Meanwhile, Brazilian supplies have continued to be available in smaller sizes, although prices have increased; this supply should be available indefinitely. These sizes are satisfactory for production of modern military crystal units, and yield figures are improving and may be expected to reach those of the end of World War II in the near future despite smaller size and poorer quality of quartz. All known potential domestic reserves have been cataloged. No competition with either Brazilian or synthetic quartz is anticipated, although the information thus derived has insurance value. The most spectacular success has been realized in the development of synthetic quartz under Signal Corps sponsorship. Crystals have been grown at reasonable rates in lots up to 50 pounds and in sizes up to 700 grams, or nearly 2 pounds. On the basis of available evidence, it will become

competitive with natural quartz on a yield basis if not a per pound basis. Preliminary results from a broad program of evaluation indicate that synthetic quartz will be a satisfactory substitute for the natural product, and its future regular use seems assured.

## STUDIES OF SILICA TRANSFER AND DIFFUSION

RICHARD G. YALMAN

ANTIOCH COLLEGE

Oxygen exchange experiments between water and dissolved silica in sodium silicate solutions and in gradient experiments using a quartz source and sodium carbonate solutions as well as in isothermal experiments using a silica glass source and both sodium chloride and sodium fluoride solutions, indicate that the soluble silica is present in a monomeric form having either a three or a four-fold coordination. In basic solutions the exchange experiments show that silica dissolves directly to form a silicate ion. Growth and devitrification experiments under isothermal conditions show that the rate of conversion of silica glass to quartz increases with increasing pH, i.e. under the conditions favorable for increasing concentrations of silicate ions. These results, together with an analysis of the structure of cristobalite, quartz and other silicates, indicate that cristobalite is formed under hydrothermal conditions from orthosilicic acid while quartz is formed from dihydrogen silicate ions. This is supported by x-ray examination of devitrified material and of silica grown on seed plates. In basic solutions only quartz is found while in neutral and weakly acid solutions the major portion of the devitrified material consists of cristobalite. Furthermore, in weakly acid solutions, cristobalite has been grown on quartz seed plates.

Appendix 5. Symposium Program, Eighth Annual Frequency  
Control Review of Technical Progress

—symposium—program—

*8th Annual*

FREQUENCY CONTROL REVIEW

of

TECHNICAL PROGRESS

12 - 14 APRIL 1954

**SIGNAL CORPS ENGINEERING LABORATORIES**

**FORT MONMOUTH, N. J.**

SIGNAL CORPS ENGINEERING LABORATORIES  
FORT MONMOUTH, NEW JERSEY

The Frequency Control Symposium Program has been prepared under the supervision of the Frequency Control Branch, Squier Signal Laboratory, and is published for the information and guidance of all concerned. Suggestions or criticisms relative to the form, contents, purpose or use of the publication should be referred to the Signal Corps Engineering Laboratories, Fort Monmouth, New Jersey, Attention: Chief, Frequency Control Branch.

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SYMPOSIUM PROGRAM

EIGHTH ANNUAL REVIEW OF TECHNICAL PROGRESS

SIGNAL CORPS ENGINEERING LABORATORIES

FREQUENCY CONTROL BRANCH

FORT MONMOUTH, NEW JERSEY

12, 13, 14 April 1954

Berkeley-Carteret Hotel, Asbury Park, N. J.

Outline of Meetings

Monday, 12 April 1954

|            |                                                                    |                 |
|------------|--------------------------------------------------------------------|-----------------|
| 9:00 a.m.  | Registration                                                       | Palm Terrace    |
| 10:30 a.m. | General Session                                                    | Crystal Terrace |
| 11:30 a.m. | Luncheon                                                           | Oval Lounge     |
| 1:30 p.m.  | Properties of Crystalline Materials and of Piezoelectric Vibrators | Crystal Terrace |
| 4:15 p.m.  | Quartz Synthesis                                                   | Crystal Terrace |

Tuesday, 13 April 1954

|            |                                                       |                 |
|------------|-------------------------------------------------------|-----------------|
| 9:00 a.m.  | Aging Forum                                           | Crystal Terrace |
| 11:45 a.m. | Temperature Control Systems                           | Crystal Terrace |
| 12:15 p.m. | Luncheon                                              |                 |
| 1:30 p.m.  | Temperature Control Systems (Contd)                   | Crystal Terrace |
| 2:00 p.m.  | Research and Development in the United Kingdom        | Crystal Terrace |
| 3:00 p.m.  | Frequency Control by Means Other Than Quartz Crystals | Crystal Terrace |
| 7:00 p.m.  | Annual Dinner                                         | Crystal Terrace |

Wednesday, 14 April 1954

|            |                                                    |                 |
|------------|----------------------------------------------------|-----------------|
| 9:00 a.m.  | Filters, Oscillators and Frequency Control Systems | Crystal Terrace |
| 12:15 p.m. | Luncheon                                           |                 |
| 1:30 p.m.  | Production Forum                                   | Crystal Terrace |

## Detailed Schedules

MONDAY MORNING, 12 April 1954

### General Session

- 9:00 to 10:30 a.m. Registration
- 10:30 a.m. Introductory Program - Signal Corps Engineering Laboratories
- Mr. W. L. Doxey, Chief, Frequency Control Branch, Squier  
Signal Laboratory
- Lt. Col. John V. Fill, Director, Squier Signal Laboratory
- Col. F. F. Uhrhane, Commanding, Signal Corps Engineering  
Laboratories
- Dr. Harold A. Zahl, Director of Research, Signal Corps  
Engineering Laboratories
- 11:30 a.m. Luncheon - Guest Speaker, J. L. Grever, Radio Corporation  
of America  
"Application of Quartz Crystals to Color Television Equipment"

MONDAY AFTERNOON, 12 April 1954

### Properties of Crystalline Materials and of Piezoelectric Vibrators

1. 1:30 p.m. Research and Development Within Frequency Control Branch -  
E. A. Gerber - Frequency Control Branch, SCEEL
2. 2:00 p.m. Lattice Parameters of Natural and Synthetic Quartz -  
D. L. Hammond - Frequency Control Branch, SCEEL
3. 2:30 p.m. Quartz Crystal Imperfections - G. W. Arnold, Jr. -  
Naval Research Laboratory
4. 3:00 p.m. X-Ray Diffraction Studies of Piezoelectric Crystals -  
R. Pepinsky - The Pennsylvania State University
- 3:30 p.m. Intermission
5. 3:45 p.m. Mathematical Theory of Vibrations of Elastic Bodies -  
R. D. Mindlin - Columbia University

### Quartz Synthesis

6. 4:15 p.m. *a* Growth of Quartz at Low Pressures - D. R. Hale -  
Brush Laboratories Company
7. 4:45 p.m. *a* Growth of Quartz Crystals at High Temperatures and Pressures -  
A. C. Walker - Bell Telephone Laboratories

### TUESDAY MORNING, 13 April 1954

#### Aging Forum

1. 9:00 a.m. *a* An Evaluation of Metals and Techniques for Coating Quartz  
Piezoelectric Resonators - R. B. Belser and W. H. Hicklin -  
Georgia Institute of Technology
2. 9:30 a.m. *e* Frequency Aging of Plated High Frequency Crystal Units -  
A. W. Warner - Bell Telephone Laboratories
3. 10:00 a.m. *e* Aging Effects on HF Crystals and VHF Crystal Units -  
P. E. Mulvihill - Frequency Control Branch, SCEL
- 10:30 a.m. Intermission
4. 10:45 a.m. *e* Study of Aging Effects on Military Plated Crystal Units -  
P. D. Gerber - Radio Corporation of America
5. 11:15 a.m. Discussion

#### Temperature Control Systems

6. 11:45 a.m. *d* Lavoie Crystal Ovens - N. E. Tetrault - Lavoie Laboratories
- 12:15 p.m. Luncheon

### TUESDAY AFTERNOON, 13 April 1954

#### Temperature Control Systems (Contd)

1. 1:30 p.m. *d* Triple Point Thermostats - C. P. Saylor and R. Alvarez -  
National Bureau of Standards

#### Research and Development in the United Kingdom

2. 2:00 p.m. Advancements in Research and Development in the United  
Kingdom - H. T. Mitchell - British Post Office

Frequency Control By Means Other Than Quartz Crystals

3. 3:00 p.m. *g* Frequency Stabilization by Non-piezoelectric Crystals -  
R. M. Gogolick and R. M. Ulmer - Horizons, Inc.
- 3:30 p.m. Intermission
4. 3:45 p.m. *g* Frequency Control Above 500 Mc - D. W. Fraser -  
Georgia Institute of Technology
5. 4:15 p.m. *h* Spectroscopic Line Breadth of Microwave Frequencies -  
R. H. Dicke - Princeton University
6. 4:45 p.m. *h* Reduction of Doppler Broadening of the Ammonia Spectral  
Line - L. E. Norton - Radio Corporation of America

TUESDAY EVENING, 13 April 1954

- 7:00 p.m. Annual Dinner - Crystal Terrace - Berkeley Carteret Hotel  
Guest Speaker - J. E. Doerr - National Park Service  
"Your National Parks and Monuments"

WEDNESDAY MORNING, 14 April 1954

Filters, Oscillators and Frequency Control Systems

1. 9:00 a.m. *L* Low Frequency Electro-Mechanical Filters - S. L. Lapin -  
Motorola, Inc.
2. 9:30 a.m. *g* Feasibility Study for Packet Oscillator Design - H. E. Gruen  
and E. A. Roberts - Armour Research Foundation
3. 10:00 a.m. *g* Crystal Oscillators in the VHF Region - G. L. Davies -  
Davies Laboratories
- 10:30 a.m. Intermission
4. 10:45 a.m. *g* Transistor Oscillators - B. J. Dasher - Georgia Institute  
of Technology
5. 11:15 a.m. *g* Present Status of Radio Frequency Control Systems in  
Military Radio Communications Equipment - R. E. Lacy -  
Radio Communications Branch, SCEL
6. 11:45 a.m. *g* The Computer Type Decade Frequency Generator - R. W. Frank -  
General Radio Company
- 12:15 p.m. Luncheon

WEDNESDAY AFTERNOON, 14 April 1954

Production Forum

1. 1:30 p.m. Crystal Requirements for Future Radio Equipments -  
R. E. Lacy - Radio Communications Branch, SCEL
2. 2:00 p.m. e Quartz Crystal Reliability Problems - W. R. Prendergast -  
Canadian Signals Research and Development Establishment,  
Canada
3. 2:30 p.m. Current Mobilization Activities - Signal Corps Supply Agency,  
Philadelphia, Pa.
- 3:00 p.m. Intermission
4. 3:15 p.m. j Process Control of Inductance in Crystal Manufacturing -  
T. G. Clark - Western Electric Company
5. 3:45 p.m. j Some Problems Encountered in the Production of High Harmonic  
Crystals - Pioneer Electric and Research Corporation
6. 4:15 p.m. j Discussion of Crystal Unit Manufacturing Techniques -  
G. F. Fisher - Midland Manufacturing Company, Inc.
- 4:45 p.m. Discussion

Appendix 6. Abstracts, Eighth Annual Frequency  
Control Review of Technical Progress

## Abstracts of Papers to be Presented

### RESEARCH AND DEVELOPMENT WITHIN FREQUENCY CONTROL BRANCH

E. A. GERBER

FREQUENCY CONTROL BRANCH, SCEL, FORT MONMOUTH

A brief description of the internal research and development program on frequency control is presented. The following items are treated in more detail:

1. Parameters of CR-18/U and CR-23/U crystal units as a function of the geometry of the crystal plate.
2. Activity dips in VHF crystal units as influenced by geometry of the blanks, the surface finish and the mounting structure.
3. Amplitude-frequency effects in VHF crystal units as a function of the mounting structure.
4. Investigation of the accuracy of measurement with the CI-Meter TS-683/TSM.

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N O T E S

## LATTICE PARAMETERS OF NATURAL AND SYNTHETIC QUARTZ

D. L. HAMMOND

FREQUENCY CONTROL BRANCH, SCEL, FORT MONMOUTH

The lattice parameters of various samples of natural and synthetic quartz have been measured as part of a program to determine the important imperfections of quartz and their relation to the characteristics of resonators prepared from different samples. It has been established that the lattice parameters are definitely a structure sensitive physical property.

A method of precision lattice parameter measurement has been developed which employs high Bragg angle parallel positions of a double crystal spectrometer. This technique provides a sensitivity of six seconds of arc for a spacing change of one part in  $10^6$  (ppm) and sharp diffraction patterns whose width is .014 that of the  $\text{CuK}\alpha_1$  line in an equivalent single crystal spectrometer. Temperature control to a few hundredths of a degree C has been maintained by a solid-liquid eutectic point. Reproducibility of the lattice parameter measurements is one-tenth ppm and the systematic errors are less than one ppm.

Variations of 40 ppm have been observed in the spacing of clear and smoky natural quartz. Studies of synthetic quartz grown by the Bell Telephone Laboratories and Brush Development Company have indicated that these crystals are homogeneous within three or four parts in  $10^6$  and have spacings similar to natural quartz. The Bell crystals have a spacing smaller than the Brush crystals by approximately 15 parts in  $10^6$ .

Although the diffraction pattern is comparatively narrow, its width and form are very sensitive to crystal imperfections. The narrowest diffractions obtained were from natural clear quartz with some of the synthetic quartz providing diffractions nearly as sharp. However, other synthetic samples display a highly asymmetrical diffraction pattern with a width over twice that of natural clear quartz. This asymmetry is due to spacing variation rather than to an orientation defect.

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N O T E S

## QUARTZ CRYSTAL IMPERFECTIONS

G. W. ARNOLD, JR.

NAVAL RESEARCH LABORATORY

Attempts to verify the existence of an amorphous surface layer on quartz have been without success. Recent investigations by Finch<sup>1</sup>, et al, however, with a 140 Kv. electron diffraction camera, have shown positive evidence for the existence of a layer approximately  $1 \text{ \AA}$  in thickness. These results will be discussed.

Investigations of the optical absorption spectra of synthetic and natural quartz have recently been initiated and preliminary results will be presented.

<sup>1</sup> Finch, Lewis, and Webb, Proc. Phys. Soc. (London), 66B, 949, 1953

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N O T E S

# X-RAY DIFFRACTION STUDIES OF PIEZOELECTRIC CRYSTALS

RAY PEPINSKY

THE PENNSYLVANIA STATE UNIVERSITY

Accurate crystal structure analyses have been carried out on normal and ammoniated Rochelle Salt (RS), ammonium dihydrogen phosphate (ADP) and cadmium niobate. The aim of the analyses is to establish the nature of the bonding systems in these crystals, the effect of temperature changes upon these, and the structural mechanisms of the phase transitions.

Rochelle Salt, which is ferroelectric in the temperature range from  $-18^{\circ}\text{C}$  to  $24^{\circ}\text{C}$ , has been subjected to very careful X-ray and neutron diffraction analyses. These reveal that the ferroelectric transitions are chiefly marked by shifts in positions of certain of the hydrogen nuclei. New systems of O-H...O bonds are found, which participate in both piezoelectric and ferroelectric activity; the earlier theories relating structure and electrical behaviour require serious modification, as a consequence. The space group of ferroelectric Rochelle Salt is  $P2_1$ , and there are actually 56 atoms in the asymmetric unit of the cell. The neutron diffraction study, which must take all these atoms into account, is thus more complicated than any complete structure analysis yet accomplished by X-ray diffraction.

The dielectric measurements of Kurtshatov, on a series of RS crystals with various degrees of substitution of  $\text{NH}_4^+$  ions for  $\text{K}^+$  ions, have been re-taken, and it is confirmed that although a small amount of  $\text{NH}_4^+$  (above 2%) destroys the ferroelectric activity of PS, substitution of from 15% to 90%  $\text{NH}_4^+$  renders the crystals ferroelectric again, at low temperatures. An X-ray study indicates that the  $\text{NH}_4^+$  ion preferentially replaces one set of  $\text{K}^+$  ions in the crystals.

Whereas  $\text{KH}_2\text{PO}_4$  (KDP) becomes ferroelectric at  $-151^{\circ}\text{C}$ , ADP becomes anti-ferroelectric at  $-125^{\circ}\text{C}$ . The structural reason for the difference in the dielectric behavior of these crystals has been sought by X-ray analyses. The ADP transition is now fully elucidated, as was that in KDP previously. Neutron studies of the KDP transition are also reported.

The crystal structure of  $\text{Cd}_2\text{Nb}_2\text{O}_7$  has been examined by single-crystal methods at room temperature and at  $-120^{\circ}\text{C}$ , since the structure becomes ferroelectric below  $-90^{\circ}\text{C}$ . Some aspects of the transition from the cubic (room temperature) structure to the rhombohedral ferroelectric structure are revealed.

A new low-temperature X-ray goniometer, for structural studies down to liquid helium temperature, is described. This is being used in a very accurate study of the nature of the bonding in quartz.

# MATHEMATICAL THEORY OF VIBRATIONS OF ELASTIC BODIES

R. D. MINDLIN

COLUMBIA UNIVERSITY

This report is a review of recent developments in the mathematical theory of vibrations of elastic bodies. It is well known that the elementary equations of vibration are suitable only for low frequencies. These include the Bernoulli-Euler equation for flexural vibrations of beams, the Bernoulli equation for longitudinal vibrations of rods, the Lagrange equation for flexural vibrations of plates, and the plane-stress equations for face vibrations of plates. On the other hand, the three-dimensional equations of the mathematical theory of elasticity are too difficult to solve for bodies having more than one finite dimension. The recent developments are systems of equations suitable for much higher frequencies, but no more difficult to solve than the elementary equations which they replace. Applications will be described to problems of: (1) coupling between thickness-shear and flexural vibrations in a strip and in a circular disk; (2) coupling between thickness-shear, thickness-twist, and flexural vibrations in a strip and in a circular disk; (3) coupling between face and thickness vibrations of a circular disk; and (4) coupling between longitudinal and radial vibrations of a rod. In addition, applications to crystalline and piezoelectric media will be described.

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N O T E S

## GROWTH OF QUARTZ AT LOW PRESSURES

D. R. HALE

## BRUSH LABORATORIES COMPANY

The Signal Corps sponsored quartz project at The Brush Laboratories Company in the past year reached full rated performance of about 80 pounds of synthetic quartz growth per month at our increased quantity quartz facility. This part of the total project has now moved from the laboratory stage to full pilot-plant status and is under continued operation under the Signal Corps Supply Agency.

The pilot-plant operates at an average temperature of 360°C and at a pressure of 5000 to 6000 pounds per square inch as previously found satisfactory. The quartz produced is of high grade in terms of optical and crystallographic inspection. Broad evaluation in terms of oscillator plate requirements is being made possible by distribution of this material to qualified manufacturers by the Signal Corps.

Research on quartz growth has continued on a reduced level of expenditure, the efforts being directed particularly (1) to clarifying further the physical-chemical basis of our process, and (2) to studying variations which may lead to greater economy.

(1) Phase relations in the quaternary system,  $\text{Na}_2\text{O}-\text{CO}_2-\text{H}_2\text{O}-\text{SiO}_2$ , were further examined. The apparent contradiction between retrograde solubility of quartz shown in a diagram last year, and increased solubility indicated by the function of our process, was resolved.

Other solutions, such as more dilute sodium carbonate, and mixtures of carbonate and hydroxide, grew quartz successfully and have certain advantages.

Special nutrient materials, silica gel granules and cracked synthetic crystals, were used in two runs; the crystals are to be particularly studied for the relative amount of impurities.

(2) Study of improved growing processes was concentrated on growth at low pressures (1000 to 2000 psi) and on comparison of the two-chamber rocking autoclave with the single vertical chamber type. Two of the latter of 14-gallon capacities were acquired and are being operated at 1200 psi. Growth rate has been under 15% of our standard rate, and is accompanied by lower costs of equipment, maintenance, and power consumption.

Another pair of single vertical autoclaves for operation up to 10,000 psi were acquired and have been used for comparison with rocking autoclaves under approximately standard growing conditions.

8,54

# GROWTH OF QUARTZ CRYSTALS AT HIGH TEMPERATURES AND PRESSURES

A. C. WALKER

BELL TELEPHONE LABORATORIES, INC.

During 1953 the work at the Bell Telephone Laboratories on the synthesis of quartz crystals, under Signal Corps Contract DA36-039 SC-5572, has been mainly on the following objectives:

## 4" Diameter Autoclaves

1. To study the phase relations in the quartz-growing system at different NaOH concentrations, and correlate with rate and quality of growth; also to determine the distribution of quartz in the system at the end of the run so as to evaluate the efficiency of the process;
2. To grow quartz crystals of good quality to weights of about one pound each, at moderate growth rates;
3. To determine if circulation by convection is adequate in the long unit so that larger numbers of smaller crystals may be grown at rapid rates, consistent with the best results obtained in smaller units;
4. To avoid corrosion due to failure of the silver plated parts in the growing chamber and thus take advantage of the superior stability of sodium hydroxide solution against spontaneous nucleation;
5. To obtain further information on growth rates vs. degree of supersaturation;
6. To study heat distribution along the length of the tubes so as to correct the condition leading to caking of the nutrient just below the baffle.

The results obtained to date on these objectives are:

It was predicted, on the basis of work of Morey and Hesselgesser, that at concentrations above 0.5 N. NaOH, two phases exist in the autoclave. One of these, a heavy liquid phase, was estimated to contain more dissolved silica than that present in the gas phase in which growth takes place. Not only would this heavy liquid phase dissolve and hold this quartz, making it unavailable for growth, but its presence might impede circulation

## GROWTH OF QUARTZ CRYSTALS AT HIGH TEMPERATURES AND PRESSURES (Contd)

of the gas phase, retarding solution and transport of dissolved quartz to the growing zone.

Experiments now completed show that these predictions are true. Quartz of equal or better quality may be grown more rapidly and efficiently from 0.5 N. NaOH solution than at higher concentrations, and there is no evidence of a heavy liquid phase under these conditions. Furthermore, calculations were made, based on our experiments, showing the amounts dissolved in each phase and the distribution of silica in the system at the end of the run. We have been able to check the total nutrient before and after a run and check within 20 grams of total amounts of either 4000 or 8200 grams.

Several one pound quartz crystals have been grown in a single run in a little over two months. These are quite free from imperfections and are extremely clear in appearance. This result is confirmation of several others, which demonstrates that the process is reproducible and that excellent quality of quartz may be grown under controlled conditions, to as large a size as may be desired consistent with equipment available.

Recent results demonstrate that circulation by convection is adequate to give high growth rates, at the same time transporting large amounts of quartz. In one recent test, nearly twice as much quartz was deposited in 30 days on 60 crystals as on a few one pound crystals in two months. In another test, 30 crystals grew to 80-160 grams each in 30 days at rates up to 0.04" per day. It is expected that 60 crystals, each weighing about 125 grams, may be grown in less than one month.

Failure of the silver plating may be avoided by not evacuating the system after filling. Spontaneous nucleation is thus avoided and temperature differences greater than 40°C, may be possible with stability of solution. It is considered that bubble imperfections may be overcome in other ways than by evacuating the system to remove air.

It appears that the following generalization may be made, the rate of growth in 0.5 N. NaOH solution, expressed in mils/day is of the same order as temperature difference in °C. For example, a 30° difference gives a rate of about 0.03" per day. It appears reasonable to predict growth rates of 0.05" per day under our operating conditions.

Regarding heat distribution, it has been established that a suitably located wrap-around heater will prevent caking of the nutrient.

9201

## GROWTH OF QUARTZ CRYSTALS AT HIGH TEMPERATURES AND PRESSURES (Contd)

### 1" Diameter Autoclaves

1. To compare rates of growth and quality of the product at lower temperatures and pressures with those obtained at the normal operating conditions;
2. To evaluate the relative merits of sodium hydroxide and carbonate, as well as mixtures of these two reagents on the growth of quartz;
3. To study the use of water glass solutions to conserve nutrient;
4. To investigate the effect of impurities in the solution on the growth of quartz;
5. To use other metals than steel as liners, so as to eliminate the need for silver plating.

Our results on this phase of the investigation has been curtailed by difficulty experienced with leaks in the top welds of the small liner tubes. One reason seems to be defects in some new steel tubing recently obtained for this work.

There is some evidence that a mixture of the carbonate and hydroxide gives faster growth rates than the hydroxide alone, and maintains adequate stability against spontaneous nucleation. Thus far the results at lower temperatures and pressures do not compare favorably with those at higher operating conditions. Water glass solution appears to contain some impurities which have a very bad effect on the quality of growth of quartz. Attempts will be made to determine what these are. No significant data are available on the remaining objectives listed above.

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N O T E S

AGING STUDY OF METAL PLATING ON QUARTZ CRYSTALS  
AN EVALUATION OF METALS AND TECHNIQUES FOR  
COATING QUARTZ PIEZOELECTRIC RESONATORS

RICHARD B. BELSER AND WALTER H. HICKLIN

GEORGIA INSTITUTE OF TECHNOLOGY

A versatile, high-rate, sputtering apparatus has been constructed with four-inch Pyrex pipe-fittings. At 20 ma, 3750 volts and a pressure of approximately  $3 \times 10^{-2}$  mm of mercury, gold and silver may be deposited at 300-500 angstroms per minute, and platinum and rhodium at 200-250 angstroms per minute.

Resistivity and structural examinations, before and after aging by heating, have been conducted for sputtered films of Pd, Pt, Rh, Ir, Ru and Os. These metals are extremely stable to changes in film structure induced by heating within a vacuum. Little evidence of grain growth of the metals in electron micrographs or diffraction patterns until temperatures near 600°C are attained. Ruthenium and osmium, however, are subject to considerable oxidation when heated in air.

Tests of the adherence of sputtered gold films to quartz substrates have been conducted. Excellent adherence was obtained by subjecting the quartz substrate to a five-minute positive-ion bombardment at 100 ma current and 3000 Volts prior to sputtering. Examination of a polished quartz surface by the electron-microscope replica-technique, after successive five-minute periods of positive-ion bombardment, showed definite micro-etching of the substrate. This etching and elimination of the adsorbed gas layer on the substrate, immediately prior to and during the sputtering action, appear to be largely responsible for the improved adhesion.

Extensive tests of sputtered gold as a coating for both polished and etched 6 mc AT-cut quartz resonators have been made. Comparative tests of indium, bakelite and Dupont No. 4817 silver cement bonds have shown that indium solders consistently gave crystals of lowest dynamic-resistance parameters and highest "Q" values.  $R_e$  values of 4 ohms and "Q" values above 200,000, for 6 mc AT-cut crystals, have been attained. A detailed account of crystal preparation, coating and mounting procedures will be given.

Resonators coated with sputtered rhodium and platinum have shown stabilities comparable to those coated with sputtered gold. For good crystals frequency drifts of less than 0.0003% per year are not unusual.

8,59

# FREQUENCY AGING OF PLATED HIGH FREQUENCY CRYSTAL UNITS

A. W. WARNER

BELL TELEPHONE LABORATORIES, INC.

A review of the measurements of frequency aging at the Bell Telephone Laboratories during the past ten years has shown a consistent improvement from about 20 PP  $10^6$ /week for crystal units in phenolic holders to 1.0 PP  $10^6$ /month for CR32 type units and to 0.01 PP  $10^6$ /month for glass enclosed, polished crystal units.

Our approach to the problem of frequency aging has been an attempt to reduce contamination and its effects. Where metal holders were used, such techniques as special cleaning of the covers and fluxless soldering materially reduced the negative aging characteristic of such units. The use of glass enclosures, which may be more effectively cleaned both before and after assembly, improved the aging characteristic. By polishing the crystal plate and by applying compact gold electrodes, the area exposed to the residual contamination was greatly reduced, resulting in still further improvement of the aging characteristic.

It is believed that by applying the principles outlined, crystal units with greatly improved aging over that obtained generally can be produced in quantity. Pertinent data will be shown by means of slides.

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N O T E S

AGING EFFECTS  
ON HIGH FREQUENCY AND VERY HIGH FREQUENCY CRYSTAL UNITS

P. E. MULVIHILL

FREQUENCY CONTROL BRANCH, SCEL, FORT MONMOUTH

Measurements were made on the changes in frequency of CR-23/U type crystal units. The crystals were aged in two ovens: a small oven, with a capacity of sixty units operated at  $75 \pm 0.2^\circ\text{C}$ ; a larger oven with a capacity of one-hundred and forty-four units operated at  $85 \pm 0.01^\circ\text{C}$ . Measurements were made on the nominal, fundamental frequency of 16.5 Mc and on the third and fifth harmonics. Twenty percent of the crystal units were pressure mounted; the remainder had gold-sputtered electrodes, were clip-mounted and bonded with four different bonding agents. Nine sub-groups of each type of bonding varied as to surface finish and time of etch. All units were vacuum baked and sealed in vacuo. Frequency measurements were made daily over a period of five weeks.

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N O T E S

# STUDY OF AGING EFFECTS ON MILITARY PLATED CRYSTAL UNITS

P. D. GERBER

RADIO CORPORATION OF AMERICA  
RCA VICTOR DIVISION

Previous crystal unit aging studies in the frequency range of 80 kc to 500 kc revealed a general trend of frequency deviation in a positive direction due to continued exposure to elevated temperatures. A practical limit of  $\pm .0005\%$  in 6 months at  $+50^{\circ}\text{C}$  was indicated. A similar investigation of higher frequency CR-19 and CR-23 units, now conducted for the Signal Corps, already has shown several interesting indications.

1. Many carefully made crystal units, supposedly fabricated and hermetically sealed to comply with MIL-C-3098 specifications, actually have minute leaks which permit contamination of the quartz plate faces and a serious negative frequency change exceeding  $.0005\%$  in 30 days at  $75^{\circ}\text{C}$ .
2. Attainment of low frequency deviations requires the utmost cleanliness of all surfaces forming the crystal cavity, and the final sealing of the holder must be in clean, dry gas if the units are not left evacuated.
3. Unplated crystals pressure air-gap mounted between recessed and plated quartz electrodes show very good frequency stability, less than  $.00002\%$  deviation in 30 days, which may be largely due to the care exercised in the cleaning and sealing operations.
4. Crystal units having polished quartz plate surfaces also show very good frequency stability, among the best of all units tested thus far in this investigation.

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N O T E S

## LAVOIE CRYSTAL OVEN

N. E. TETRAULT

### LAVOIE LABORATORIES

The Lavoie Crystal Oven was originally developed for use in the two frequency meters produced by Lavoie, the FR-5 and the FR-6. Performance of the oven surpassed by far that which was required or expected. As a result, the oven is now produced as an electronic component and sold to other equipment manufacturers.

The oven consists of a crystal chamber, two thermostats and two heating elements. One of the thermostats is a small bi-metallic type and controls current through a heating element which is used only for quick warm-up. This thermostat opens a few degrees below the operating temperature. The other thermostat is larger and, by virtue of its size and construction, is considerably more precise in its operation. Its expanding element is a thin-walled brass tube which is hermetically sealed and houses the lever mechanism and contacts. The lever mechanism is externally adjustable to set the operating temperature. This thermostat controls the current in the other heating element, which maintains the oven at its operating temperature. The thermal mass and the insulation of the oven is at the maximum allowed by the requirements of small size, ambient temperature range and precise temperature control.

This oven is available with operating temperatures of 75 degrees or 35 degrees Centigrade, plus or minus one degree. The total temperature variation of a given oven at room temperature will be plus or minus 0.025 degrees C, and less than 0.15 degrees C over the ambient temperature range of -40 to plus 70 or 80 degrees C, depending upon the oven operating temperature.

The warm-up time from -25 degrees C is approximately 7 minutes. Units are manufactured which operate from 6, 24, or 115 volts, AC or DC, with an operating power consumption of 15 watts (30 watts during fast warm-up). The oven will plug into a standard octal socket and is 4" high, 1 7/8" wide, and 1 7/16" deep. It will take any crystal in crystal holders type HC-6 or HC-13. (A different model oven is supplied for the two types of holders.)

Since this oven was designed primarily for military use, it meets many of the military environmental requirements.

A method of electronic temperature measurement called the Electronic Differential Temperature Meter has been developed whereby small temperature variations can be measured and recorded with an Esterline-Angus recording meter. Each thermostat is checked with the Electronic Differential Temperature Meter for operating temperature accuracy, temperature excursion (peak to peak operating temperature variation) and contact performance. The ovens are well aged and re-checked to insure that each oven meets its high standards of precision and reliability.

## TRIPLE-POINT THERMOSTATS

CHARLES PROFFER SAYLOR AND ROBERT ALVAREZ

### NATIONAL BUREAU OF STANDARDS

The triple point of a chemical substance is the temperature and pressure combination at which solid, liquid, and vapor coexist. For a perfectly pure substance, the temperature under these conditions is definite. It is the aim under this program of study to develop a temperature-stable oven to hold crystal oscillators. Surveys of available chemical compounds have indicated that para-dibromobenzene has desirable properties: a triple point at the right temperature, adequate rate of melting or crystallization, comparatively low price, chemical stability, sufficient latent heat of fusion, etc.

The design and construction of ovens has been such that the desired stability has been presently achieved but not with adequately small size or with sufficiently automatic or fool-proof manipulation. To operate successfully the chemical substance must ballast the temperature by its own melting and freezing. It must be well isolated thermally so that the system will never depart very much from total phase equilibrium. There must be a sensing device so that roughly the same amount of the compound will be kept melted at all times during operation. The choice of this volume operated sensing device together with its proper operation constitutes the most pressing problem at this time. Design of a device for control of warm-up has not been attempted yet but it does not seem likely that it will be difficult to manage. Thermal isolation should be improved over that which has been achieved so that it will be feasible to use smaller and lighter equipment.

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N O T E S

ADVANCEMENT IN RESEARCH AND DEVELOPMENT IN THE UNITED KINGDOM

H. T. MITCHELL

BRITISH POST OFFICE

1. Quartz growing.
2. Cutting of quartz.
3. Overtone crystals.
4. Precise frequency standards.
5. Buried standards.
6. Synthesis of frequencies.
7. Filters.
8. Frequency control in the microwave range.

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N O T E S

# FREQUENCY STABILIZATION BY NON-PIEZOELECTRIC CRYSTALS

R. M. GOGOLICK AND R. M. ULMER

HORIZONS INCORPORATED

This paper describes a system for frequency stabilization based on an electrostatic type of excitation which shows excellent possibilities for commercial usage as frequency controllers or narrow band filter elements in the medium range of performance and price. The control element is a single crystal or polycrystalline metal rod or sphere vibrating in a particular mechanical resonance mode. The vibrations can be excited and detected electrostatically and the resultant voltage fed back to the driving oscillator. Q's ranging from several hundred to over 120,000 have been found in the frequency range from 10 to 500 ac. An excellent fine adjustment of the resonators to frequency is obtainable.

For constant frequency over a temperature range the resonator material must satisfy the condition that the negative of the thermoelastic coefficient equal the coefficient of linear expansion over the desired range. The development of two materials, one in the form of a single crystal, and the other polycrystalline, is described. The polycrystalline material employed in a bread-board oscillator with a 12AX7 tube operated within the tolerance of the frequency counter used ( $\pm 0.1$  c.p.s. at a nominal frequency of 46,171.8 c.p.s.) for a period of 15 hours with no temperature control employed. Measurements over a temperature range from  $-40^{\circ}\text{C}$  to  $+90^{\circ}\text{C}$  showed it to be superior to commercial quartz. This resonator also had excellent stability with respect to variations in power supply voltage.

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N O T E S

## FREQUENCY CONTROL ABOVE 500 MC

D. W. FRASER

### GEORGIA INSTITUTE OF TECHNOLOGY

The recent development of UHF television and the present emphasis on other electronic UHF systems by military agencies has brought about strict requirements for the control of the frequencies involved. Early developments of frequency-control methods were designed specifically to apply to certain discrete frequency bands. These are now being supplemented by various means of providing a general coverage of a wide range of the ultrahigh-frequencies. A brief survey of documented control methods applicable at frequencies above 500 Mc shows that several novel developments have now been employed. These include various forms of the familiar butterfly tuner, a split-cylinder resonator, a capacitor with distributed inductance, and a microwave discriminator circuit.

With the possible exception of the last named, none of these methods provide basically the frequency stability necessitated in present applications. Hence attention is being directed toward the use of cavities of various configurations as elements for precision control of frequency. For frequencies below 1500 Mc the coaxial cavity provides the only attractive configuration. On this project primary emphasis has been directed toward design and construction of coaxial cavities of suitable characteristics. The maximization of impedance and Q and the minimization of temperature sensitivity have been primary objectives. Temperature insensitivity has been sought by a process of compensation as well as by utilization of small-expansion materials such as fused quartz, Invar, or Stupalith.

Precise frequency control can be effected if the stable characteristics of the cavity can be applied directly to an associated oscillator. Unfortunately, considerable difficulty is experienced in finding satisfactory methods of coupling the oscillator to the cavity. Four distinct applications of the cavity have been tested or are under consideration. These include utilization of the cavity as (1) a two-terminal reactance shunted across interelectrode capacities, (2) a series impedance in a feedback loop, (3) a source of shock-excited voltage driving an oscillator or regenerative amplifier, and (4) as the frequency-determining element in an AFC system. In the latter case it may be utilized as the frequency-determining element in a true discriminator system or may act as a phase-determining element as one terminal of a microwave bridge. In any and all cases the cavity has excellent inherent capabilities of providing precision frequency control.

# SPECTROSCOPIC LINE BREADTH OF MICROWAVE FREQUENCIES

R. H. DICKE

PALMER PHYSICAL LABORATORY  
PRINCETON UNIVERSITY

It is sometimes convenient to divide the causes of the breadth of a spectroscopic line into three classes, motional breadth, collision breadth, and radiation breadth.

The various collision mechanism which lead to a contribution to line breadth are discussed. The effects of wall and gas collisions on motional breadth are described. Natural radiation breadth which is usually ignored is shown to be of considerable importance in some cases.

\* \* \* \* \*

N O T E S

# REDUCTION OF DOPPLER BROADENING OF THE AMMONIA SPECTRAL LINE

L. E. NORTON

RCA LABORATORIES DIVISION

Need for ever improved constancy of time and frequency standards is basic. The quartz control element has been highly developed and is used in the best clocks and frequency standards. However, it now appears that further great improvements in quartz standards are unlikely. Some different mechanism is required, and is at hand. Frequencies which are precisely defined by "ordinary" microwave spectral lines are being used to control the frequencies of oscillators.

The observed spectral lines all depend on transitions between discrete energy levels of elementary particles, ordinarily molecules or atoms. Spectral line frequencies, used as frequency standards, are free from secular drift to a remarkable degree. Bandwidths of these "ordinary" lines are about  $9 \times 10^4$  at  $2.4 \times 10^{10}$  cycles/second, and are due to a number of causes. Most important are particle to particle collisions, particle to absorption cell wall collisions, quantum mechanical spectral line flattening due to excessive excitation field, and the Doppler contribution. Methods for reducing the bandwidth contributions of all except that due to the last named cause have been known for a long time. Until recently, reduction of the Doppler component has been a major obstacle.

In a noise free environment, a frequency is defined with perfect precision, excluding secular drifts, by a spectral line of any bandwidth. However, fundamental noise is always present in the world in which we live. Therefore, for spectral lines of equal intensities, the precise frequency information in a narrower bandwidth line is less obscured and degraded by the noise. To achieve any significant improvement in controlled-frequency stability, methods had to be devised for obtaining narrow bandwidth spectral lines of good intensity, and control systems which made effective use of the precise frequency information in these lines had to be produced. Obviously, the required worthwhile reduction in spectral line bandwidth could only be achieved by a material reduction of Doppler contribution to total bandwidth. A good method for Doppler reduction is defined loosely as one in which the race between bandwidth reduction and accompanying loss in spectral line intensity is won by bandwidth reduction.

A special absorption cell containing a mechanism for producing Doppler reduced bandwidth spectral lines, and a system for utilizing effectively the precise frequency information in these narrow bandwidth lines, were designed and constructed. Salient features and some preliminary results will be described.

## LOW FREQUENCY ELECTROMECHANICAL FILTERS

S. P. LAPIN

MOTOROLA, INC.

The use of filters as frequency selective elements has become of increasing importance with the development of the electronics art, and their use has become standard in the intermediate frequency sections of certain types of communication receivers. Electromechanical filters have several advantages over the corresponding conventional inductance-capacitance filters, including a selectivity curve with a flatter acceptance nose, steeper rejection slopes, and lower insertion loss. In addition, size and weight are reduced in an electromechanical filter, and higher orders of stability can be maintained. The filters can be sealed and permanently adjusted in the factory.

Two main types of electromechanical filters are feasible. The first uses a piezoelectric or a magnetostrictive element as an equivalent two terminal electrical network, which network is utilized in conjunction with other electrical elements to form the filters. The second method depends on converting electrical energy to mechanical energy, utilizing the mechanical motion characteristics to determine the bandpass, and then reconvert the mechanical energy into electrical energy. Usually the converting elements at both ends of the structure are either piezoelectric or magnetostrictive.

Theory and experimental data on these two main general types of filters are presented.

\* \* \* \* \*

N O T E S

# FEASIBILITY STUDY FOR PACKET OSCILLATOR DESIGN

H. E. GRUEN - E. A. ROBERTS

## ARMOUR RESEARCH FOUNDATION

A study to determine the feasibility of designing a standard series of packaged crystal oscillators meeting the majority of Air Force equipment requirements is reported. Information is tabulated for Air Force equipment currently in development and production employing crystals for frequency control. This information is then used to establish oscillator requirements for specifying a series of packaged oscillators that will find greatest use in future equipment design.

Thirty-nine crystal oscillator circuits are catalogued; these circuits comprise four specific circuit types and a few special application circuits. Data obtained for each circuit include crystal type, operating frequency, stability, output voltage, crystal drive level, and operating conditions peculiar to each circuit.

Three basic oscillators are proposed for the standard series. These are the Colpitts, Electron Coupled Colpitts, and the Cathode Coupled oscillator circuits. The first two circuits, operating with anti-resonant crystals, will provide respectively low and high impedance sources and cover the frequency range up to sixteen megacycles. The third circuit will be used in the standard packet series at frequencies above sixteen megacycles.

This circuit study has shown that at least 70 per cent of all oscillator requirements of present equipments could be met with the proposed series. In future equipments, through proper selection of operating frequencies in synthesizing networks, it should be possible to specify standard packaged oscillators in 80 to 90 per cent of all application.

\* \* \* \* \*

N O T E S

# CRYSTAL OSCILLATORS IN THE VHF REGION

G. L. DAVIS

THE DAVIES LABORATORY, INC.

This report will present the results of studies of the performance characteristics of series mode, overtone crystal oscillator circuits operating in the 75 to 135 mc frequency range. Four different circuits have been investigated: transformer-coupled, C.I. meter, cathode-coupled, and bridged-T. Data will be presented showing frequency stability and correlation, output, crystal power, and other operating variables, in graph and comparison chart form.

Applicability of these circuits to operational equipment will be discussed and the relative advantages and disadvantages of each pointed out. Problems involved in tuning and ganging such circuits over a wide frequency range are somewhat difficult, but by no means insurmountable. Both capacitive and inductive tuning methods have been explored; ganged variable inductors appear desirable, but pose severe problems if other tuned circuits (such as later stages of a transmitter) must be ganged with the crystal oscillator.

Buffer design has been standardized, as it has been found that the buffer should be considered as an integral part of the oscillator in critical applications.

A brief summary of crystal admittance data will be given, and problems connected with crystal power measurement in operating circuits will be outlined.

\* \* \* \* \*

N O T E S

# TRANSISTOR OSCILLATORS

B. J. DASHER

GEORGIA INSTITUTE OF TECHNOLOGY

Fundamental considerations in transistor oscillators are essentially the same as for vacuum-tube oscillators. The same basic circuits may be used in each case. There are, however, substantial differences in detailed requirements which lead to a different approach to design problems, especially in the case of point-contact transistor oscillators. Some of the difficulties that arise are discussed in this paper. Among these problems may be mentioned impedance matching, amplitude control, internal phase shift and bandwidth considerations, and parasitic oscillations. Emphasis is placed on point contact transistors and the relative advantages of certain prototype circuits are pointed out.

\* \* \* \* \*

N O T E S

PRESENT STATUS OF RADIO FREQUENCY CONTROL  
SYSTEMS IN MILITARY RADIO COMMUNICATION EQUIPMENT

RAYMOND E. LACY

RADIO COMMUNICATION BRANCH, SCEL, FORT MONMOUTH

The importance of frequency control systems is emphasized by the demands for more and more communication channels in an already overcrowded radio-frequency spectrum. Existing and contemplated types of control systems are shown in their relation to operational requirements, and in their relation to piezoelectric crystal units. It is shown that there is still much room for engineering ingenuity in applying such techniques, although many large scale investigations have been conducted by all military services in the field of frequency control during the last decade. The Signal Corps has lead the way in achieving the most universal equipments from a frequency control viewpoint. Typical equipment are shown, and their frequency determining portions described.

\* \* \* \* \*

N O T E S

# THE COMPUTER TYPE DECADE FREQUENCY GENERATOR: PROGRESS IN 1953

R. W. FRANK

GENERAL RADIO COMPANY

Since the first description of the decade-frequency generator at this symposium last year, a working three-decade system using a high speed scale-of-"N" divider has been constructed and tested. The basic principles of this system are reviewed.

Performance data obtained for the model is discussed and limitations are discovered and evaluated. Problems of noise and stability dictate the addition of an automatic tuning system and improvements to the phase detector.

The range of the experimental model has been extended to cover frequencies between 1-10 Mc and the techniques for accomplishing this end are described and evaluated.

Consideration of the foregoing limitations and improvements now points to definite ultimate limitations on the range of operation of the final model. It is concluded that a six-decade system to cover the frequency range to 10 Mc in 10 c. steps is theoretically possible with the improvements described.

\* \* \* \* \*

N O T E S

## CRYSTAL REQUIREMENTS FOR FUTURE RADIO EQUIPMENTS

RAYMOND E. LACY

RADIO COMMUNICATION BRANCH, SCEL, FORT MONMOUTH

Radio transmission-medium characteristics and equipment functions are described as they bear on future piezoelectric oscillator and filter crystal requirements. Temperature stability, power-drain limitations in heaters, and miniaturization are the important variables. The most stringent temperature characteristics are in the long range frequencies (approximately 3 mc/sec to 30 mc/sec). Here basic stabilities of the order of one part in ten to the ninth power is operationally needed. Extreme miniaturization is the key to portable, short-range radio sets in the tactical frequencies (approximately 20 mc/sec to 100 mc/sec). The 0.005% stability crystal units may be utilized here, but the design engineer could provide additional desirable simplicity in operation and maintenance as a result of any further order of crystal stability that is achievable without appreciably increasing the economic cost of the crystals. Such features are incorporated into the equipment by utilizing more of the crystal temperature tolerance to compensate for wider tolerances in the remainder of the frequency determining electronic circuits and systems. In the "long lines" radio systems known as radio relay the frequencies (50 mcs to 10,000 mcs or higher) are obtained by "spectrum generators," or other more conventional multipliers which are actuated by over-tone crystals. In the field of crystal filters new requirements have arisen from the better understanding provided by application of communication theory. These characteristics are discussed as they affect future crystal needs.

\* \* \* \* \*

N O T E S

# QUARTZ CRYSTAL RELIABILITY PROBLEMS

W. R. PRENDERGAST

## CANADIAN SIGNALS RESEARCH AND DEVELOPMENT ESTABLISHMENT

This paper deals with the wire-mounted, metal plated quartz crystal as employed in the solder sealed metal holder. Quantities of the crystal unit type CR-23/U have been used in connection with the production of a portable radio set for the Canadian Army. After a period of storage, a high failure rate was noted although the units passed source inspection at the time of manufacture. The Canadian production procedure disclosed the fact of failure.

A group known as the Quartz Crystal Panel was formed. It was composed of Services representatives. The aim of this panel was to improve the quality of quartz crystal units and to encourage designers to take full advantage of the characteristics of the crystal. The rest of this paper discusses the research and development program, outlining tests, test procedures and results.

\* \* \* \* \*

N O T E S

SOME PROBLEMS ENCOUNTERED IN THE PRODUCTION OF  
HIGH HARMONIC CRYSTALS

PIONEER ELECTRIC AND RESEARCH CORPORATION

A description will be given of some of the work done in devising methods and controls for measuring the physical and electrical characteristics of the crystals, with comment on the significance of the results of these efforts.

\* \* \* \* \*

N O T E S

Appendix 7. Symposium Program, Ninth Annual Frequency  
Control Review of Technical Progress

# **SYMPOSIUM PROGRAM**

## **9th ANNUAL**

### **FREQUENCY CONTROL REVIEW**

#### **OF**

### **TECHNICAL PROGRESS**

**25, 26, 27 MAY 1955**



**SIGNAL CORPS ENGINEERING LABORATORIES**  
**FORT MONMOUTH, N. J.**

SIGNAL CORPS ENGINEERING LABORATORIES  
FORT MONMOUTH, NEW JERSEY

The Frequency Control Symposium Program has been prepared under the supervision of the Frequency Control Branch, Components Division, and is published for the information and guidance of all concerned. Suggestions or criticisms relative to the form, contents, purpose or use of the publication should be referred to the Signal Corps Engineering Laboratories, Fort Monmouth, New Jersey, Attention: Chief, Frequency Control Branch.

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SYMPOSIUM PROGRAM

NINTH ANNUAL REVIEW OF TECHNICAL PROGRESS

SIGNAL CORPS ENGINEERING LABORATORIES

FREQUENCY CONTROL BRANCH

FORT MONMOUTH, NEW JERSEY

25, 26, 27 May 1955

Hotel Berkeley-Carteret, Asbury Park, N. J.

Outline of Meetings

Wednesday, 25 May 1955

|            |                                                                       |                 |
|------------|-----------------------------------------------------------------------|-----------------|
| 8:30 a.m.  | Registration                                                          | Palm Terrace    |
| 10:15 a.m. | General Session                                                       | Crystal Terrace |
| 11:30 a.m. | Luncheon                                                              |                 |
| 1:00 p.m.  | Properties of Crystalline Materials<br>and of Piezoelectric Vibrators | Crystal Terrace |

Thursday, 26 May 1955

|            |                                                   |                 |
|------------|---------------------------------------------------|-----------------|
| 9:00 a.m.  | Stability Characteristics in<br>Frequency Control | Crystal Terrace |
| 12:15 p.m. | Luncheon                                          |                 |
| 1:30 p.m.  | VHF and UHF Frequency Control                     | Crystal Terrace |
| 3:45 p.m.  | Quartz Synthesis                                  | Crystal Terrace |
| 7:00 p.m.  | Annual Dinner                                     | Crystal Terrace |

Friday, 27 May 1955

|            |                                  |                 |
|------------|----------------------------------|-----------------|
| 9:00 a.m.  | High Precision Frequency Control | Crystal Terrace |
| 12:15 p.m. | Luncheon                         |                 |
| 1:30 p.m.  | Production Forum                 | Crystal Terrace |

Detailed Schedules

WEDNESDAY MORNING, 25 May 1955

General Session

8:30 to 10:15 a.m. Registration

10:15 a.m. Introductory Program - Signal Corps Engineering  
Laboratories

Dr. E. A. Gerber, Chief, Frequency Control Branch,  
Components Division

Lt. Colonel John V. Fill, Director, Components Division

Brig. General F. F. Uhrhane, Commanding, Signal Corps  
Engineering Laboratories

Dr. Harold A. Zahl, Director of Research, Signal Corps  
Engineering Laboratories

11:30 a.m. Luncheon

WEDNESDAY AFTERNOON, 25 May 1955

Properties of Crystalline Materials and of Piezoelectric Vibrators

1. 1:00 p.m. Research and Development in Frequency Control -  
E. A. Gerber - Frequency Control Branch, SCEL
2. 1:30 p.m. Mathematical Theory of Vibrations of Elastic Plates -  
R. D. Mindlin - Columbia University
3. 2:00 p.m. Determining Strain Patterns in Thickness Shear  
Resonators - K. S. Van Dyke - Wesleyan University
4. 2:30 p.m. Frequency Spectra in Quartz Resonators - C. R. Miggins  
and A. D. Frost - Tufts College
- 3:00 p.m. Intermission
5. 3:15 p.m. Effects of Ionic Diffusion on the Physical Properties of  
Quartz - H. E. Wenden - Harvard University
6. 3:45 p.m. Optical Absorption Spectra Studies of Natural and Synthetic  
Quartz - G. W. Arnold, Jr. - Naval Research Laboratory
7. 4:15 p.m. Effects of Impurities on the Resonator and Lattice  
Properties of Quartz - Donald L. Hammond - Frequency  
Control Branch, SCEL

THURSDAY MORNING, 26 May 1955

Stability Characteristics in Frequency Control

1. 9:00 a.m. *n* An Evaluation of Metals and Techniques for Coating Quartz Piezoelectric Resonators - Richard B. Belser and Walter H. Hicklin - Georgia Institute of Technology
2. 9:30 a.m. *e* Study of Aging Effects on Military Plated Crystal Units - P. D. Gerber - Radio Corporation of America
3. 10:00 a.m. *d* Frequency-Temperature Behavior of Resonators of Natural and Synthetic Quartz - Rudolf Bechmann - The Brush Laboratories Company
- 10:30 a.m. Intermission
4. 10:45 a.m. *e* Methods of Obtaining Reduced Tolerances in Crystals Over a Wide Temperature Range - L. F. Koerner - Bell Telephone Laboratories, Inc.
5. 11:15 a.m. *g* Transistor Oscillators - E. Gonzales-Correa - Solid State Devices Branch, SCEL
6. 11:45 a.m. Research and Development in Canada - L. F. Bennett, Canadian Military Electronics Standards Agency, and D. M. Eisen, Canadian Radio Manufacturing Company
- 12:15 p.m. Luncheon

THURSDAY AFTERNOON, 26 May 1955

VHF and UHF Frequency Control

1. 1:30 p.m. *g* Packet Oscillator Circuits - E. Roberts and H. Gruen - Armour Research Foundation
2. 2:00 p.m. *n* VHF Crystal Test Circuits - G. K. Guttwein - Frequency Control Branch, SCEL
3. 2:30 p.m. *n* Methods of Measuring the Equivalent Electrical Parameters of Quartz Crystals - William B. Wrigley - Georgia Institute of Technology
- 3:00 p.m. Intermission
4. 3:15 p.m. *g* Precision Frequency Control Above 500 MC - Donald W. Fraser and Edward G. Holmes - Georgia Institute of Technology

### Quartz Synthesis

5. 3:45 p.m. *a* Laboratory and Pilot Plant Growth of Quartz at Moderate Pressure - D. R. Hale, Hans Jaffe, W. H. Charbonnet - The Brush Laboratories Company
6. 4:15 p.m. *a* Growth of Quartz at High Temperatures and Pressures - A. C. Walker - Bell Telephone Laboratories, Inc.

### THURSDAY EVENING, 26 May 1955

7:00 p.m. Annual Dinner - Crystal Terrace, Hotel Berkeley-Carteret - Guest Speaker - Willy Ley - "A Survey of Space Satellite Proposals"

### FRIDAY MORNING, 27 May 1955

#### High Precision Frequency Control

1. 9:00 a.m. *f* The Primary Frequency and Time Standard - W. D. George - National Bureau of Standards
2. 9:30 a.m. *f* High Precision Crystal Measurements - A. W. Warner - Bell Telephone Laboratories, Inc.
3. 10:00 a.m. Research and Development in the United Kingdom - H. T. Mitchell - British Post Office Engineering Department
- 10:30 a.m. Intermission
4. 10:45 a.m. *h* Molecular and Atomic Frequency and Time Standards - F. H. Reder - Frequency Control Branch, SCEI
5. 11:15 a.m. *h* Spectroscopic Line Breadth of Microwave Frequencies - R. H. Dicke - Princeton University
6. 11:45 a.m. *h* An Atomic Frequency Standard - Jerrold R. Zacharias - Massachusetts Institute of Technology
- 12:15 p.m. Luncheon

FRIDAY AFTERNOON, 27 May 1955

Production Forum

1. 1:30 p.m. VHF Crystal Resonators - E. M. Shideler - Scientific Radio Products, Inc.
2. 2:00 p.m. Methods of Measurement and Test of Crystal Units in Britain - W. J. Young - Standard Telephone and Cables Ltd.
3. 2:30 p.m. High Stability Crystal Units - Otis Ivie - James Knights Company
4. 3:00 p.m. Current Mobilization Activity - Arnold Ratner - Signal Corps Supply Agency
- 3:30 p.m. Intermission
5. 3:45 p.m. Production Procedures for VHF Crystals (5th Harmonic, 60-100 MC) - R. D. Cortright - Union Thermoelectric Corporation
6. 4:15 p.m. Ultrasonic Quartz Cutting - Norman E. Gibbs - Raytheon Manufacturing Company
7. 4:45 p.m. Mechanization of Crystal Manufacturing Processes - Lester V. Wise - Bulova Research and Development Laboratories, Inc.
- 5:15 p.m. Discussion

Appendix 8. Abstracts, Ninth Annual Frequency  
Control Review of Technical Progress

# RESEARCH AND DEVELOPMENT IN FREQUENCY CONTROL

E. A. GERBER

## SIGNAL CORPS ENGINEERING LABORATORIES

A description of the research and development program on frequency control is presented. It is shown that most of the contractual and internal projects of Frequency Control Branch are based on the following requirements for frequency control devices:

1. Improved control of parameters.
2. Better stability.
3. Improved frequency-temperature characteristics.
4. Extension to higher frequencies.
5. Miniaturization.

The progress made in the last year is reviewed and results of research and development on selected projects are given.

\* \* \* \* \*

N O T E S

# MATHEMATICAL THEORY OF VIBRATIONS OF ELASTIC PLATES

R. D. MINDLIN

During the past year, several advances have been made in the mathematical theory of vibrations of plates which has been under development by the group at Columbia.

On the theoretical side, the equations have been extended in scope and, at the same time, placed on a firmer foundation by devising a formal procedure for passing from the three-dimensional theory to plate theory. The technique adopted is an expansion of all the variables and equations of the three-dimensional theory in an infinite power series in the thickness coordinate. The subsequent process of truncating the series leaves no doubt as to the nature of the approximations inherent in each order of approximation. The process also brings to light the equations governing the face and thickness modes, their coupling with each other and with the thickness-shear and flexural modes.

Several new applications have been executed. All of the problems of thickness-shear and flexural vibrations of rectangular plates, which had previously been solved only for isotropic plates, have now been solved for crystal plates. The phenomenon of coupling at a free edge is now better understood as a result of recognizing the theoretical significance of the simply supported edge, at which no coupling occurs, and studying solutions of the equations for elastically supported edges, which include the free and simply-supported edges as limiting cases.

The equations governing the coupled face and thickness vibrations of isotropic plates, which were described last year, have been studied further. The solution for the case of coupled radial and thickness vibrations of a circular disk has been examined in detail and has been found to conform very well with experiments made with barium titanate disks at Bell Telephone Laboratories last summer.

There is under preparation an extensive monograph describing in detail all of the developments and results obtained to date.

In addition, work has been started on an extension of the theory to the VHF range and also on the incorporation of the electric and thermal properties in the present form of the general theory.

# DETERMINING STRAIN PATTERNS IN THICKNESS SHEAR RESONATORS

K. S. VAN DYKE

WESLEYAN UNIVERSITY

After a period of mistrust of the Mindlin analytical treatment of thickness shear resonators, arising in a failure to understand the intent of the analysis, we have during the past two years been using Mindlin's final equations as our guide in the experimental exploration of vibration patterns in quartz plates. We are finding good qualitative agreement between theory and experiment, and have thus far no hint of any quantitative discrepancy either. The analysis in question certainly provides an insight into the details of modes which theory had not theretofore included.

Our experiment uses a small probe electrode and readings of the magnitude of local admittance per elemental area taken at many different locations over the square face of an AT-cut quartz plate of given dimensional ratio, thus surveying local variations of response at the driving frequency. A single resonance is surveyed at a time and the admittance is plotted as a sort of contour map for that mode. A number of such maps have been compared with strain patterns computed from Mindlin's surface amplitude distribution function for the x-direction. These modes have been positively identified, among the host of modes excited by such small electrodes, by the agreement in both long-wave "carrier" and ripple as to the number of half waves which are included within the x-dimension of the plate. The spreading of the field from the small probe tends to mask the full swing of the ripple pattern when the ripple "wave" is short and introduces some limitation on our ability to obtain a precise measure of the ratio of amplitude of "carrier" to ripple for a quantitative comparison with theory on this point.

In our experiment the probe is connected into the mode-analyzer and the latter is set for a very slow sweeping rate. Following a technique initiated by Hok in a very early report the deflection of the beam for the mode under investigation is noted in each position of the exploring probe.

# FREQUENCY SPECTRA IN QUARTZ RESONATORS

C. R. MINGINS AND A. D. FROST

TUFTS COLLEGE

In the work thus far we have found that in some regions of frequency-dimension space the two-dimensional theory of Mindlin for rectangular plates gives an excellent check with experiment, but that in other regions the agreement is not very good. In a severely rectangular piezoid the multiplicity of mode families have their maximum opportunity to be driven, and all together present a very confusing array of modes. At the same time they offer an unusual opportunity for a study of the different varieties and their interplay, if one is in a position to select the desired ones for critical examination. The combination of the probe technique, selective segment plating, and inspection of the coupling evidence together yield a very powerful method of mode identification. We are attempting to obtain thickness-shear modes of increasingly higher secondary order.

The shaping of electrodes as a method for producing a "clean" spectrum gives considerable promise. We are attempting to make the shape conform more closely to the actual distribution of amplitude over the plate surfaces as revealed by probing; this should have salutary effects upon the spectrum. The photographic method of applying shaped electrodes in proper alignment is a versatile one and should make it possible to get the utmost out of the mode suppression technique.

Methods are discussed for studying the modes which assume greater importance at higher drive levels.

\* \* \* \* \*

N O T E S

# OPTICAL ABSORPTION SPECTRA STUDIES OF NATURAL AND SYNTHETIC QUARTZ

G. W. ARNOLD, JR.

NAVAL RESEARCH LABORATORY

The optical absorption spectra of x-irradiated clear natural quartz crystals from several localities, smoky quartz, amethyst and citrine have been examined. The colored varieties have been studied in their natural state and when recolored by x-irradiation after thermal bleaching. Several new ultraviolet bands in amethyst have been observed. The evidence for and against the current hypotheses for the mechanism of coloration of quartz and fused silica will be presented and some conclusions offered.

The spectra of American and British synthetic quartz has been studied in both the ultraviolet and the near infrared. Significant differences are observed in the location of the main ultraviolet absorption maximum in synthetic quartz as compared to natural quartz. Differences in absorption at 2.92  $\mu$  have also been noted and a possible explanation will be presented.

\* \* \* \* \*

N O T E S

# EFFECTS OF IMPURITIES ON THE RESONATOR AND LATTICE PROPERTIES OF QUARTZ

DONALD L. HAMMOND

## SIGNAL CORPS ENGINEERING LABORATORIES

An investigation has been conducted to determine the effects of imperfections upon the properties of quartz. The properties investigated were the frequency-temperature characteristics of quartz resonators and the lattice parameters. The material investigated in this study includes synthetic quartz grown upon AT seed plates from sodium carbonate solutions containing separately the impurities aluminum, germanium, lead, boron, selenium, titanium, tin, calcium and zirconium. Quartz synthesized by the Brush Laboratories Company on CT and Y bar seeds, by Bell Telephone Laboratories on CT seeds at high temperatures and pressures and by the British Post Office on Z cut seeds were also investigated.

The synthetic material grown by various laboratories under differing conditions has been fabricated into piezoelectric resonators of the AT thickness-shear type. Measurements have been made of the frequency-temperature characteristics of the fifth mode at 30 mc over the temperature range from  $-65$  to  $+125^{\circ}\text{C}$ . Significant variations have been observed in the resonator properties of the synthetic material relative to natural quartz. The effects of impurities are substantial and desirable in several cases.

Lattice parameter measurements of the synthetic material indicate that under similar growing conditions the material is quite homogeneous. Variations observed in the lattice spacing from different samples of Brush synthetic quartz are smaller than the variations observed in the spacing of various samples of natural quartz. The addition of the impurities aluminum and germanium produces a substantial strain in the crystal as indicated by the increase in the diffraction pattern width. The width of the diffraction pattern taken from the aluminum and germanium doped quartz is several hundred seconds compared with a diffraction pattern width of 20 to 40 seconds for the normal synthetic material. In general, the effect of these two impurities is to increase the lattice spacing as much as 70 parts per million over the nominal value for synthetic material.

# AN EVALUATION OF METALS AND TECHNIQUES FOR COATING QUARTZ PIEZOELECTRIC RESONATORS

RICHARD B. BELSER AND WALTER H. HICKLIN

GEORGIA INSTITUTE OF TECHNOLOGY

The adherence of both evaporated and sputtered films are improved by positive-ion bombardment just prior to deposition. Substrate temperatures of 400°C were measured during bombardment and the residual heat, in addition to aiding adherence, partially pre-ages the film as deposited. The adherence of evaporated gold and aluminum films was increased from 100 to 300 psi and from 200 to 600 psi, respectively. Changes in films, including numbers of pin holes and bubbles, normally produced by artificial aging, were reduced. Metals of the transition series gave high adherence, 1650 psi being obtained for sputtered nickel films.

Frequency studies of 175 additional 6-10 mc resonators have emphasized destabilizing influences attributable to the metal plating and to fabrication techniques. The frequencies of coated crystals are affected by changes of mass or by changes of the moment inertia of the vibrating system. Additions of mass are caused by corrosion and adsorption; losses of mass by desorption, evaporation of bonding cement components and ejection of nonadherent particles. Changes in the moment of inertia of the system are caused by alloying of bimetal films, bonding solder or cement diffusion and by recrystallization of the metal film. Concurrently, small changes in electrical parameters occur but their frequency effects are masked by the moment of inertia changes.

Corrosion may be avoided by the choice of gold, rhodium or platinum as the plating metal. Adsorption and desorption may be limited by meticulous cleaning, mounting and hermetic sealing procedures, using a dry inert gas near a pressure of one atmosphere as the filler. Alloying affects, and the accompanying moment of inertia changes, can be limited or eliminated respectively by careful matching of bonding solders or cements with the metal plating and by refusal to use bimetal films. Recrystallization effects for rapidly sputtered films, or for evaporated aluminum films, are small or absent at crystal operation temperatures. Resonators coated with sputtered gold or platinum and bonded with Du Pont No. 5806 silver cement, and sputtered rhodium, bonded with indium, have given outstandingly stable performance. Coatings of evaporated aluminum bonded with indium or silver cement, although slightly inferior to the above metals, give excellent stability on polished crystals.

# STUDY OF AGING EFFECTS ON MILITARY PLATED CRYSTAL UNITS

P. D. GERBER

RADIO CORPORATION OF AMERICA

Crystal units exhibit a frequency change with time, a characteristic generally referred to as aging. In an effort to evaluate the effect of variations of fabrication on aging, approximately 100 CR-23 crystal units were subjected to aging tests at 75°C. The variations in fabrication included gas filling with nitrogen, helium, or dry air, base plating by evaporation with and without additional plating to frequency, and base plating by sputtering. Some of the units were "pre-aged" by storing for 24 hours at 125°C. The effect of pre-aging lowered the frequency, but the majority of units so treated showed no marked improvement in aging over those not pre-aged. Aging is aggravated when units are frequency adjusted by additional plating. Another group of crystal units fabricated with covers annealed in a hydrogen atmosphere had a tendency to age upward in frequency in contrast to the downward aging observed with untreated covers. Upward aging was also observed with covers cleaned with sodium cyanide, and suggests inadequately cleaned covers as a source of contamination. Units of relatively low aging were deliberately punctured to produce a leak. This immediately produced a pronounced downward frequency aging. After gas filling and resealing, the frequency aging followed the original characteristic but usually at a lower frequency than before puncturing. The effect on frequency of mounting stresses introduced by loop misalignment was also determined. In addition, crystal units made on IPS contracts are included in this aging study.

\* \* \* \* \*

N O T E S

# FREQUENCY-TEMPERATURE BEHAVIOR OF RESONATORS OF NATURAL AND SYNTHETIC QUARTZ

RUDOLF BECHMANN

THE BRUSH LABORATORIES COMPANY

Natural quartz from different sources has displayed remarkable uniformity as far as all piezoelectric applications are concerned. Regardless of the source of electronic-grade quartz used, when the crystallographic orientation of a piezoelectric resonator plate is specified, no significant variations are encountered in the performance of the resulting resonator plates.

Investigations into the frequency-temperature behavior of AT-type quartz crystals have revealed that differences between natural and synthetic quartz exist. The angle of orientation at which the minimum frequency change for a given temperature range occurs is slightly different, and small differences have been found in the frequency-temperature curves for corresponding angles. To describe the frequency-temperature behavior analytically, the measured change of frequency versus temperature can be developed in a power series, determined by first, second, and third order temperature coefficients. In the temperature range from  $-60$  to  $+100^{\circ}\text{C}$ , higher order temperature coefficients can be neglected. For AT-type crystals the maximum and minimum temperature, the corresponding total frequency change, and the inflection temperature are characteristic quantities. The three temperature coefficients and their variation with the angle of orientation describe the family of frequency-temperature curves of AT-type resonators in the vicinity of the angle giving the zero temperature coefficient. The three temperature coefficients are functions of the orientations and to some extent of the dimensional ratio of the plate; the type of mounting also has some influence. The coefficients vary with the order of overtones. For AT-type resonators made from natural quartz and synthetic quartz grown under various conditions, these coefficients have been determined for certain cases.

It has been recently found possible to modify the properties of synthetic quartz by introducing some other elements during the growing process. An example is quartz grown in an alkaline solution containing germanium dioxide. Measurements have been made on AT-type resonators cut from synthetic quartz modified by germanium addition. The temperature coefficients for these resonators have also been determined. The third order temperature coefficient for the AT-type resonator is found noticeably reduced and as a consequence of this, the frequency-temperature curves are flattened over a wider temperature range. Compared with natural quartz, the inflection temperature is shifted to higher values.

METHODS OF OBTAINING REDUCED TOLERANCES IN CRYSTALS  
OVER A WIDE TEMPERATURE RANGE

L. F. KOERNER

BELL TELEPHONE LABORATORIES, INC.

This paper describes methods of compensating the frequency change with temperature of a crystal unit by means of thermistor networks. The networks, in effect, add a reactance in series with the crystal unit, the reactance being varied by means of one or more thermistors so as to compensate for frequency changes with temperature of the crystal unit. Examples showing the networks as used in transistor and vacuum tube oscillators are given.

The degree of compensation obtained for several typical crystal units employing some of the networks is shown. Proposed circuit and outline arrangements are offered which would be expected to meet the Services' requirements for several of the standard crystals now procured under Specification C-3098 for operation under temperature control.

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N O T E S

## TRANSISTOR OSCILLATORS

E. GONZALEZ-CORREA

### SIGNAL CORPS ENGINEERING LABORATORIES

The design and operation of free-running and crystal-controlled transistor oscillators appear in some ways to be an extension of vacuum tube practice. Additional complexity of design arises as a result of attempts to represent transistor physics at high frequencies in terms of an equivalent electrical circuit. This approximate representation is quite sensitive to ambient, bias conditions and manufacturing tolerances. Appropriate analysis leads to the nature of the important high frequency parameters and their dependence on operating conditions. Certain design criteria follow from these results. A review of the transistor development program indicates that devices with useful high frequency oscillator properties are becoming available or have reached advanced stages of development. The properties are compared.

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N O T E S

## RESEARCH AND DEVELOPMENT IN CANADA

L. F. BENNETT

CANADIAN MILITARY ELECTRONICS STANDARDS AGENCY

D. M. EISEN

CANADIAN RADIO MANUFACTURING COMPANY

Research and development of quartz crystals in Canada during the past year has been initiated both by the Government and by industry. The responsibility for the Government-sponsored component research and development in Canada rests with the Defence Research Board and particularly the Electronics Components Development Committee and its committees. This organization has, in the past year, dealt with leak problems connected with the HC-6/U holder, general aging of plated quartz crystals, and development of a ruggedized design of plated units suitable for severe military usage.

The aging and stability problems which have been experienced with crystal units in the HC-6/U metal holders have led to the development, by the Canadian Radio Manufacturing Corporation, Toronto, Ontario, of an evacuated glass holder replacement. The glass unit is an exact physical replacement for the metal HC-6/U holder and electrical tests to date show considerably improved performance.

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N O T E S

## VHF CRYSTAL TEST CIRCUITS

G. K. GUTTWEIN

### SIGNAL CORPS ENGINEERING LABORATORIES

An investigation of the performance of Crystal Impedance Meter TS-683/TSM is presented and the upper frequency limit of the average TS-683 is established in terms of crystal resistance and power dissipation. It is shown that the TS-683 can be used for frequencies as high as 125 mc provided the existing grid current meter is replaced by one having greater sensitivity. For an extension of the frequency range beyond 125 mc, circuits with higher gain are required. A two-tube C.I. Meter is described consisting of an oscillator stage and a grounded grid amplifier. This circuit is satisfactory for frequencies up to 170 mc, if the crystal resistance does not exceed 100 ohms. Since this circuit uses the substitution method which is employed in all C. I. Meters, an evaluation of the accuracy of this method is made. The measurement error introduced by the reactive component of the substitution resistors is investigated and the deviations of frequency and resistance from their true values are given as functions of the crystal parameters and of the phase angle of the substitution resistor.

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N O T E S

# METHODS OF MEASURING THE EQUIVALENT ELECTRICAL PARAMETERS OF QUARTZ CRYSTALS

WILLIAM B. WRIGLEY

GEORGIA INSTITUTE OF TECHNOLOGY

The limitations on phase angle characteristics of substitution resistors presently used in the C. I. Meter are established as a function of crystal  $Q$  and crystal environment in the C. I. Meter circuit. Acceptable phase characteristics have been determined to exist in fixed resistor between 60 and 150 ohms only. Further limitation in the use of substitution resistors are the necessity for interpolation and the large number of resistors required.

A VHF rheostat has been developed which exhibits phase angle characteristics comparable to fixed resistors. Reactance compensation has been incorporated such that only a few units are required to cover the range of 10 to 200 ohms from 75 to 200 megacycles with acceptable phase characteristics for the C. I. Meter.

The availability of suitable VHF rheostats provides the opportunity to incorporate a null balancing bridge in a suitable oscillator for the measurement of crystal parameters. Such an instrument utilizing a two tube line-coupled oscillator is being developed and is described in detail.

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N O T E S

# PRECISION FREQUENCY CONTROL ABOVE 500 MC

DONALD W. FRASER AND EDWARD G. HOLMES

GEORGIA INSTITUTE OF TECHNOLOGY

There are two methods by which cavities may be made in order to render them virtually insensitive to temperature variations. One of these methods utilizes the compensation principle. Coaxial cavities are constructed with an extra section which withdraws the inner conductor from the cavity by the same amount by which it expands due to a rise in temperature. These types may be almost perfectly compensated at one frequency. Cavities whose temperature sensitivity is less than 0.5 ppm/°C have been fabricated.

The second method of construction utilizes materials of low expansivity such as Invar, Vycor, and Stupalith. In these types the problems of obtaining a good adhering surface and mechanical bonds became manifest. These cavities may be made tunable and exhibit excellent orders of temperature insensitivity.

Either of these cavity types may be arranged in many novel ways in order to control the frequency of an UHF oscillator. Some of these methods include a Series-Capacitor configuration, a Re-entrant type, and a Loop-Coupled type. Tests to determine the overall performance of these oscillators reveal very excellent orders of stability in the presence of ambient temperature and anode voltage variations.

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N O T E S

LABORATORY AND PILOT PLANT GROWTH OF QUARTZ  
AT MODERATE PRESSURE

THE BRUSH LABORATORIES COMPANY

D. R. HALE

New equipment put into successful operation included two vertical welded-pipe autoclaves of 1.95 cu. ft. volume. Runs in these autoclaves demonstrated a successful growth of quartz at 1200 psi and 285°C. Other new equipment included a torsion-oscillated drive for a 0.21 cu. ft. vertical autoclave, and a hydraulically operated wrench for opening and closing this size vessel.

Special seed orientation study was continued. The chosen cuts were parallel to atomic planes of moderately low indices. Some of these planes accept synthetic growth at higher rates than the natural faces or the Z-cut.

Crystals grown on the plates of various orientations and on the new Y-bar seed have in some instances exhibited flawed growth which has been especially studied. Control of this seems assured through application of presently-known information in respect to quartz growing.

HANS JAFFE

Under laboratory conditions substantially no deposition of silica takes place on the prism faces of the quartz crystal. The maximum cross section of the grown quartz crystal is therefore set by the size of the seed. For large crystals, this has required large and expensive seed plates. The cross section in the basal plane can, however, be built up if we start out from a seed bar provided this bar includes a substantial angle, preferably 30°, with the prismatic faces. It is thus possible to grow a crystal which is large in three dimensions from a seed which is large in only one dimension. In practice the crystal bar seeds cut at 30° to the prism faces (Y-bar seeds) are not grown into a crystal of the maximum basal cross section, but only to a crystal bar of optimum cross section for cutting oscillator plates.

LABORATORY AND PILOT PLANT GROWTH OF QUARTZ AT MODERATE PRESSURE  
(Contd)

W. H. CHARBONNET

Operation of the Pilot Plant for one year under a Signal Corps Industrial Preparedness Study contract was successfully concluded last September 30. The crystals produced under this program were all grown on minor rhomb seeds.

Towards the end of this period, Pilot Plant personnel cooperated with research on initial trials of the new Y-bar seed. These small scale experiments showed that the Y-bar held promise for economy both in regard to seed costs and yield of grown quartz. A new preparedness contract was therefore initiated to evaluate the process on a pilot plant scale.

In order to obtain the necessary seed material, several large, high quality stones were purchased, because the economic advantage of the Y-bar increases with the length. All subsequent seeds have been cut from plant production.

A higher rocking angle, for increased circulation, and a greater degree of filling seem to have been the key changes necessary for the growing of good Y-bar crystals.

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N O T E S

# GROWTH OF QUARTZ AT HIGH TEMPERATURES AND PRESSURES

A. C. WALKER

BELL TELEPHONE LABORATORIES, INC.

Studies of the growth of quartz hydrothermally at relatively high pressures and temperatures under Signal Corps Contracts DA36-039 SC-5572 and DA36-039 SC-64493 have resulted in several observations which suggest that a significant increase in growth rate is possible. These observations are as follows:

1. The solubility of silica in dilute alkali solutions increases with both temperature and pressure.
2. At temperatures above the critical temperature of water, transportation and growth of quartz appears to take place in a gaseous medium in which completely adequate circulation of the nutrient solution is provided by convection and turbulence.
3. Increasing the length of the autoclave simplifies the control problem.
4. Restriction of the area of the openings in the baffle between the dissolving and crystallization zones from 50 to 5% increases growth rates.
5. High temperature appears to accelerate growth rate possibly through the speeding up of intermediate reactions.

Experiments designed to take advantage of these observations have resulted in an increase in rate of growth from 30 to approximately 45 mils per day. Because of the favorable effect of this increase on the economics of the process the objectives of the contract were modified to include eight repetitive runs intended to establish the reproducibility of the process. The average growth rates obtained in this series of runs thus far is 45 mils. With the exceptions of runs terminated because of leaks no failures have been experienced.

Relative growth rates have been established approximately on various seed plates as follows: y cut < 0.1; r face 1; z face 4; z cut > 10. The high and erratic growth rate on z cut plates results in faulty growth unless

## GROWTH OF QUARTZ AT HIGH TEMPERATURES AND PRESSURES (Contd)

artificially retarded. This result suggests that a compromise is possible which will result in growth rates higher than 50 mils per day through the use of z cut seed plates grown under conditions designed to restrict growth rate and improve circulation.

Three groups of crystals were grown in which the additives germanium, aluminum, iron and selenium were introduced in the crystal in an attempt to alter crystal properties.

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N O T E S

# THE PRIMARY FREQUENCY AND TIME STANDARD

W. D. GEORGE

## NATIONAL BUREAU OF STANDARDS

The U. S. primary frequency standard agrees precisely with the mean solar second from the U. S. Naval Observatory. Observations which determine standard time extend over long intervals (weeks) before and after a particular time. Provisional frequency and time values for broadcasting are obtained from past and predicted performance of quartz crystal resonators and oscillators. Provisional values are later replaced by absolute values which are published. The performance of a precision quartz crystal unit, under controlled conditions, is such that its frequency may be precisely predicted. A prediction may be in error by a few parts in  $10^9$  for a one year interval.

The primary frequency standard is broadcast from the National Bureau of Standards' radio stations WWV and WWVH. This paper will discuss efforts and results toward improving (1) the quartz crystal clocks (2) the standard frequency broadcast services (3) the methods of adjusting and steering the broadcasts.

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N O T E S

# HIGH PRECISION CRYSTAL MEASUREMENTS

A. W. WARNER

BELL TELEPHONE LABORATORIES, INC.

The basic problem in the comparison of two frequencies to a high order of precision, e.g. one part per billion, is to overcome the effects of noise and phase jitter. Although systems relatively free of distortion have been developed for use in the laboratory, practical systems for field use are not common. The troubles likely to be encountered will be discussed, and one system using readily available components will be described.

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N O T E S

# RESEARCH AND DEVELOPMENT IN THE UNITED KINGDOM

H. T. MITCHELL

## BRITISH POST OFFICE ENGINEERING DEPARTMENT

A general survey is made of the most recent developments in connection with cultivated quartz, extreme economy in cutting quartz, overtone crystals up to 150 mc/sec and the synthesis of frequencies from a single stable source.

High precision frequency control is dealt with in more detail. Particular reference is made to Z-cut ring oscillators operating at 55°C, a Z-cut ring resonator at 12°C, the Lea resonator servo and the N. P. L. cesium resonator. Finally, details are given of the stability of the received frequency at long distances on the Rugby transmissions at 16 and 60 kc/sec.

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N O T E S

# MOLECULAR AND ATOMIC FREQUENCY AND TIME STANDARDS

F. H. REDER

## SIGNAL CORPS ENGINEERING LABORATORIES

There are two major problems to be resolved in order to establish the design of a practical atomic frequency standard. The first, and perhaps most important, is that of providing a microwave spectral line which is both sharp and strong in order that its invariant center frequency may be employed for locking a microwave oscillator having high intrinsic short-time stability. The second problem is the determination of means for dividing, or otherwise lowering, the stabilized microwave frequency to a final output frequency which may be utilized for practical applications. The method selected must not degrade the frequency stability achieved at the microwave input frequency.

Reduction in the width of spectral lines may be based upon one of three methods: Broadening due to Doppler phenomenon may be compensated by the Stark effect as in the Dicke-Norton grid cell system. A sharper line may also be obtained by a reduction of the effective thermal velocity of the radiating or absorbing molecules, either by cooling the gas or by selecting only the slow molecules, or by having the molecules collide with neutral gas particles. Finally, a reduction may be obtained by increasing the electro-magnetic wavelength of the radiation, as seen by the molecules, by using either a cavity resonator close to cut-off or by poisoning the walls of the cavity so that only neutral collisions with the walls will occur.

The increase both of the  $Q$  and the signal-to-noise ratio of the microwave spectral line has been the main target of past research. Various methods have been developed for both purposes, and  $Q$  values of more than 40 million have been attained. According to the form in which the molecules are used, the methods may be classified as being either of the gas cell type, or of the molecular or atomic beam type; the essential difference being the detection system. The increase of the signal-to-noise ratio is either based on a so-called pumping method or on a focusing method. In both cases, the ratio of the number of molecules being in the excited energy state and that being in the unexcited state can be changed in the desired direction.

# SPECTROSCOPIC LINE BREADTH OF MICROWAVE FREQUENCIES

R. H. DICKE

PRINCETON UNIVERSITY

It seems to be very important if not essential that an "atom clock" employ an atomic or nuclear resonance with an intrinsic frequency width which is very small. The one factor affecting this line width most difficult to modify is the Doppler effect resulting from atomic thermal velocities. The hyperfine transition in the monatomic vapor of the alkali metals is characterized by the fact that the Doppler effect can be essentially eliminated in a very simple fashion. If the alkali metal vapor at a pressure of perhaps  $10^{-6}$  mm of Hg is carried in a helium atmosphere at a pressure of perhaps 1 mm, the Doppler effect contribution to the line width is given by the time required by the alkali metal atom to diffuse a wavelength, rather than the time to move a wavelength as in the normal Doppler effect, an improvement of about a factor of 100. It is essential that the electronic spin orientation in the metal atom be unaffected by a collision with a helium atom. The technique of "optical pumping" for improving signal-to-noise will be discussed, also techniques for coherent pulse-induced radiation.

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N O T E S

# AN ATOMIC FREQUENCY STANDARD

JERROLD R. ZACHARIAS

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

The hyperfine structure resonance line of atomic Cesium at approximately 9192.632 mc/sec has been used to stabilize the frequency of a microwave signal generator. This resonance frequency, describable as the Larmor precession frequency of the valence electron of atomic Cesium in the magnetic field of the atomic nucleus, is observed by the atomic beam magnetic resonance method as a resonance curve with a half-width of approximately 200 cycles/sec. The techniques necessary to make it into a practical device have become available only in the last few years in connection with researches on rare isotopes of the alkali atoms. These techniques will be described.

Preliminary results show that the short-time stability thus obtained (for times less than one second) is better than 1 part in  $10^9$  and that the average stability for long times will be considerably better than this. The beam tube has run satisfactorily for periods as long as 50 days indicating that a sealed-off tube with a life of several years should be quite possible.

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N O T E S

## VHF CRYSTAL RESONATORS

E. M. SHIDELER

SCIENTIFIC RADIO PRODUCTS, INC.

A practical method has been developed for the production of VHF crystal resonators in the range from 75 mc to and including 100 mc. Regular production equipment was adapted for the purposes of lapping and polishing these crystals. The development work was done and the pilot run quantities produced using as few modifications of standard equipment as practicable in order that the process may be put to use in any crystal plant using standard equipment with a minimum of modification.

Polishing was accomplished by covering 4-inch pin-lap plates with a vinylite plastic and a method was developed for producing spiral ventilating grooves in the surfaces of these plates.

Many tests were made to try to determine what degree of flatness and/or parallelity of the surfaces is required to produce crystals that are free from activity dips over the temperature range. Also determinations were made as to the required degree of polish necessary to produce crystals of sufficiently low equivalent resistance.

Special plating patterns and spotting techniques were developed to eliminate activity dips and the work done along this line seems to indicate that the weight and distribution of the plating materials are of prime importance in the elimination of activity dips.

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N O T E S

# HIGH STABILITY CRYSTAL UNITS

OTIS IVIE

JAMES KNIGHTS COMPANY

This paper describes the problems involved in the design, production, and testing of high stability crystal units. Results of tests made on 1 mc/sec G-12A crystal units designed and manufactured by the James Knights Company are given.

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N O T E S

## CURRENT MOBILIZATION ACTIVITY

ARNOLD RATNER

### SIGNAL CORPS SUPPLY AGENCY

The Quartz Crystal Mobilization Planning Program, reactivated in October 1953, following the Korean emergency, has made great strides in its development. The responsibility for mobilization planning in the area of Quartz Crystals has been assigned to the Crystal Coordinator, Signal Corps Supply Agency. The group directly responsible to the Crystal Coordinator has maintained close liaison with all technical and mobilization authorities in the field of Quartz Crystals in order to integrate all facets of the defense activity into one overall program. Examples of work accomplished to date is the establishment of sources of supply for VHF Crystal Units up to 100 mc, initiation of a development project for mechanized equipments to produce Quartz Crystals, and maintenance of a production development organization (Union Thermoelectric Company) for the benefit of the military and industry. The needs of the other Services have been included within the planning program in the areas of equipment development and Industrial Preparedness. It is the feeling of the military that the critical nature of Quartz Crystal procurement has not lessened due to the expiration of the emergency, and it is felt that the Mobilization Planning Program will be further intensified.

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N O T E S

PRODUCTION PROCEDURES FOR VHF CRYSTALS  
(5TH HARMONIC, 60-100 MC)

R. D. CORTRIGHT

UNION THERMOELECTRIC CORPORATION

Orientation and sawing is by a simplified pre-orientation process, requiring minimum operator skills and no orientation adjustments at the sawing positions themselves, and dependent upon a system of precise jigs and fixtures.

To a thickness of 3.7 mils for the higher frequency units, the lapping process is planetary. Final lapping and polishing is with drill-press type laps at different speeds, and polishing is with plastic faced plates. In all lapping processes maximum attention is paid to abrasive and abrasive vehicle, to the conditioning of the plates, and to the carrier materials. No machine, otherwise desirable, has yet been found which has any tendency to create shapes which are beyond the surprisingly wide tolerances for flatness, parallelism, and symmetry. Avoidance of cracks, fractures, and scratches is more important. An extremely high optical polish is not absolutely essential. Control of the final lapping processes is the major problem of the entire process.

Cleaning is extremely critical, both for the crystals themselves, and for the cans and holders, to insure good activity and good aging characteristics. For the crystals a supersonic method together with vacuum drying is employed.

Aluminum plating gives better activity and freedom from activity dips, and the aging characteristics appear to be good if the aluminum is properly applied and the cans are properly cleaned and sealed. A frequency plate of sputtered gold over evaporated aluminum appears to be practical.

The conventional music wire mount with Bakelite-silver bonding, at best, adds over 5 ohms to the apparent or measured dynamic resistance, and is very erratic in its characteristics. A new mount, with solid pins and an indium bond reduces the 5 ohms to 1 ohm or less, and is conspicuously more consistent in behavior. Laboratory tests have been made.

All processes to final lapping and polishing can be performed by operators with a minimum of training and skill.

PRODUCTION PROCEDURES FOR VHF CRYSTALS (5TH HARMONIC, 60-100 MC)  
(Contd)

Meeting the military specifications for the CR-54 on up to 100 mc units, with yields comparable to or a little better than the overall industrial average, is not difficult, despite the fact that the factors which determine the difference between a 22 ohm and a 40 ohm crystal are very obscure.

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N O T E S

## ULTRASONIC QUARTZ CUTTING

NORMAN E. GIBBS

RAYTHEON MANUFACTURING COMPANY

Apparatus has been designed and built employing sonic energy, at frequencies above the upper limits of audibility, for cutting quartz crystal wafers. The processes involved in the generation of ultrasonic vibrations in a magnetostrictive nickel stack and their transmission to the actual cutting elements are briefly reviewed and the theory of cutting is described.

Application of ultrasonic principles to quartz cutting poses a number of design problems. The extremely high acceleration to which tools are subjected and the stresses created in the process create mechanical and bonding problems for which practical solutions were difficult to obtain. A number of these are discussed as contributing to understanding of the equipment developed.

Because it has been found possible to make twenty or more cuts simultaneously, it is impossible to resort to sample cutting and subsequent correction of the apparatus on the basis of X-ray examination of the wafer. The specialized problems in orientation procedure for ultrasonic cutting are presented and the techniques devised are examined.

Experimental results of ultrasonic cutting are examined in the light of requirements for highly accurate blanks cut to about one-third the thickness of those from commercial diamond saw processes. Finally, improvements required to make the apparatus developed into a practical production tool are briefly presented.

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N O T E S

# MECHANIZATION OF CRYSTAL MANUFACTURING PROCESSES

LESTER V. WISE

BULOVA RESEARCH AND DEVELOPMENT LABORATORIES, INC.

The mechanization program was initiated in response to a specific need for high production of certain crystal types immediately upon Mobilization Day.

The plant that is being designed and built by Bulova will be able to manufacture 200,000 CR-18 and CR-23 crystal units per month on a single-shift basis. It will require a greatly reduced labor force, with the training time of all employees below supervisory level limited to a maximum of two weeks. The plant is being designed to be capable of operating at capacity and shipping substantial quantities of finished units several weeks after M-Day. A substantial reduction in the price per crystal unit is expected over units made by standard methods.

As a basis for design of specific equipments, an operational analysis is being conducted for the entire plant to establish optimum man-machine combinations, maximum plant flexibility consistent with simple machine designs, and to assure proper quality and production controls. The results of this analysis to date clearly indicate the path which must be followed for successful completion of the entire development task.

The manufacturing process which has been established for mechanization differs in several respects from processes commonly used in industry today, as it was necessary to eliminate certain common processes which would result in bottlenecks in the mechanized plant. This specialized process is being established on an experimental basis in coordination with the equipment design.

Several equipments are now in the breadboard stage and have already proven the soundness of the basic designs. Others are still in experimental stages. The goal is to have all prototype equipments in operation by June 1957.

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| Deputy for Science & Technology<br>Office, Asst Sec Army (R&D)<br>(1) Washington, DC 20310                                                                                                      | Cdr, Marine Corps Liaison Office<br>ATTN: AMSEL-LN-MC<br>(1) Fort Monmouth, NJ 07703-5033 |
| HQDA (SARDA-TR)<br>Dr. Richard Chait<br>(1) Washington, DC 20310                                                                                                                                |                                                                                           |
| Director<br>Naval Research Laboratory<br>ATTN: Code 2627<br>(1) Washington, DC 20375-5000                                                                                                       |                                                                                           |
| USAF Rome Laboratory<br>Technical Library, FL2810<br>ATTN: Documents Library<br>Corridor W, STE 262, RL/SUL<br>26 Electronics Parkway, Bldg 106<br>Griffiss Air Force Base<br>(1) NY 13441-4514 |                                                                                           |
| Dir, ARL Battlefield<br>Environment Directorate<br>ATTN: AMSRL-BE<br>White Sands Missile Range<br>(1) NM 88002-5501                                                                             |                                                                                           |
| Dir, ARL Sensors, Signatures,<br>Signal & Information Processing<br>Directorate (S3I)<br>ATTN: AMSRL-SS<br>2800 Powder Mill Road<br>(1) Adelphi, MD 20783-1197                                  |                                                                                           |
| Dir, CECOM Night Vision/<br>Electronic Sensors Directorate<br>ATTN: AMSEL-RD-NV-D<br>(1) Fort Belvoir, VA 22060-5806                                                                            |                                                                                           |
| Dir, CECOM Intelligence and<br>Electronic Warfare Directorate<br>ATTN: AMSEL-RD-IEW-D<br>Vint Hill Farms Station<br>(1) Warrenton, VA 22186-5100                                                |                                                                                           |