

ARMY RESEARCH LABORATORY



# Temperature Dependent Microwave Absorption in Selected Nitrates and Nitrites

Steven G. Cornelison  
Arthur Gauss, Jr.  
U.S. ARMY RESEARCH LABORATORY

J. Krane  
J. R. Hardy  
UNIVERSITY OF NEBRASKA

ARL-TR-1179

August 1996

19961106 140

APPROVED FOR PUBLIC RELEASE; DISTRIBUTION IS UNLIMITED.

DTIC QUALITY INSPECTED 1

## **NOTICES**

**Destroy this report when it is no longer needed. DO NOT return it to the originator.**

**Additional copies of this report may be obtained from the National Technical Information Service, U.S. Department of Commerce, 5285 Port Royal Road, Springfield, VA 22161.**

**The findings of this report are not to be construed as an official Department of the Army position, unless so designated by other authorized documents.**

**The use of trade names or manufacturers' names in this report does not constitute indorsement of any commercial product.**

REPORT DOCUMENTATION PAGE			Form Approved OMB No. 0704-0188	
Public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington, VA 22202-4302, and to the Office of Management and Budget, Paperwork Reduction Project(0704-0188), Washington, DC 20503.				
1. AGENCY USE ONLY (Leave blank)		2. REPORT DATE August 1996	3. REPORT TYPE AND DATES COVERED Final, Jan 93 - Sep 95	
4. TITLE AND SUBTITLE Temperature Dependent Microwave Absorption in Selected Nitrates and Nitrites			5. FUNDING NUMBERS PR: 1L161102AH43	
6. AUTHOR(S) Steven G. Cornelison, Arthur Gauss, Jr., J. Krane,* and J. R. Hardy*				
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) U.S. Army Research Laboratory ATTN: AMSRL-WT-WD Aberdeen Proving Ground, MD 21005-5066			8. PERFORMING ORGANIZATION REPORT NUMBER ARL-TR-1179	
9. SPONSORING/MONITORING AGENCY NAMES(S) AND ADDRESS(ES)			10. SPONSORING/MONITORING AGENCY REPORT NUMBER	
11. SUPPLEMENTARY NOTES * University of Nebraska.				
12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.			12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words)  Molecular dynamics studies performed at the University of Nebraska have predicted that microwave activity should be present in some ionic molecular crystals and that significant dielectric losses may occur at or near order/disorder transition temperatures. Temperature-dependent centimeter reflection studies at frequencies from 2 to 20 GHz were performed at the U.S. Army Research Laboratory (ARL) on powdered samples of potassium nitrate, potassium nitrite, sodium nitrate, and sodium nitrite. The measurements, which show significant dielectric losses in some cases, are compared to calculations based on a Debye relaxation model. Losses observed in the salts containing nitrite ions are expected to be due to the presence of permanent dipole moments that are excited to hopping modes as the temperature is raised. Losses observed in potassium nitrate are expected to be due to the development of "induced" dipole moments as the material nears its order/disorder transition temperature of 128° C.				
14. SUBJECT TERMS molecular crystals, millimeter, wave reflection, absorption			15. NUMBER OF PAGES 20	
			16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION OF THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL	

INTENTIONALLY LEFT BLANK.

## ACKNOWLEDGMENTS

The authors would like to thank Dr. Thomas Kottke and Richard A. McGee for their timely and helpful comments while reviewing this manuscript. We would also like to thank Dr. Robert Bossoli for his expertise with the network analyzer and his help with the experimental measurements.

INTENTIONALLY LEFT BLANK.

## TABLE OF CONTENTS

	<u>Page</u>
ACKNOWLEDGMENTS .....	iii
LIST OF FIGURES .....	vii
1. INTRODUCTION .....	1
2. EXPERIMENT .....	1
3. THEORY .....	3
4. RESULTS AND DISCUSSION .....	6
5. SUMMARY .....	14
6. REFERENCES .....	15
DISTRIBUTION LIST .....	17

INTENTIONALLY LEFT BLANK.

## LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1.	Powder sample heater .....	2
2.	Phase II representation of potassium nitrate lattice .....	5
3.	Temperature-dependent microwave reflection curves for sodium nitrate powder sample .....	6
4.	Experimental reflectometer measurements and theoretical calculations for potassium nitrite at various temperatures .....	8
5.	Experimental reflectometer measurements and theoretical calculations for sodium nitrite at various temperatures .....	9
6.	Experimental reflectometer measurements and theoretical results from room temperature to 116° C for almost fully dense pressed powder potassium nitrate .....	10
7.	Experimental reflectometer measurements and theoretical results from 118° C to 136° C for almost fully dense pressed powder potassium nitrate .....	11

INTENTIONALLY LEFT BLANK.

## 1. INTRODUCTION

The dielectric properties of materials at very high frequencies (i.e., optical frequencies) and at relatively low frequencies (megahertz or less) have been studied extensively. Fewer studies have been performed within the intermediate region of centimeter- or millimeter-wave frequencies (Megaw 1973; Von Hippel 1954). Here the results of centimeter-wave reflection measurements and theoretical modeling studies for potassium nitrite ( $\text{KNO}_2$ ), potassium nitrate ( $\text{KNO}_3$ ), sodium nitrate ( $\text{NaNO}_3$ ), and sodium nitrite ( $\text{NaNO}_2$ ) are reported. Experimental studies were performed from 2 to 20 GHz in a centimeter-wave reflectometer over a temperature range from room temperature ( $25^\circ\text{C}$ ) to about  $250^\circ\text{C}$ . The dielectric losses in these materials are expected to be mostly dipolar in the frequency range studied.

The losses in the nitrites are expected to be due to the permanent dipole moments associated with the nitrite ions, which are excited to "hopping" modes when the temperature is raised (Megaw 1973; Jona and Shirane 1962). Sodium nitrate, which contains no permanent dipole moments, shows absorptions similar to that expected for ionic salts (Cornelison et al. 1992). Potassium nitrate shows interesting, temperature-dependent absorption in this frequency range. This is believed to be due to a dynamically induced dipole moment that begins to develop at temperatures just below the order/disorder transition temperature of  $128^\circ\text{C}$  (Lu and Hardy 1990, 1991; Jona and Shirane 1962).

The mechanisms that underlie the dielectric behavior of these materials are discussed in section 3. Reflection losses using a Debye relaxation model calculation are compared to and are consistent with temperature-dependent centimeter-wave reflection measurements on sodium nitrate, potassium nitrite, and potassium nitrate.

## 2. EXPERIMENT

Powder samples were measured using the powder sample heater as diagrammed in Figure 1. These salts were first powdered in an analytical mill, oven dried for several hours at  $135^\circ\text{C}$ , and then lightly packed to a thickness of a centimeter or so immediately before the measurements were to be taken. The "lightly packed" powders had approximately 50% of theoretical density. The temperature-controlled heating element is located at the bottom of the sample holder below a 0.25-in aluminum plate, which also served as a conducting back plane. Temperature variations over the back plane were typically less than  $1^\circ\text{C}$ . As the temperature was raised, the thermocouples located at various depths in the powder sample

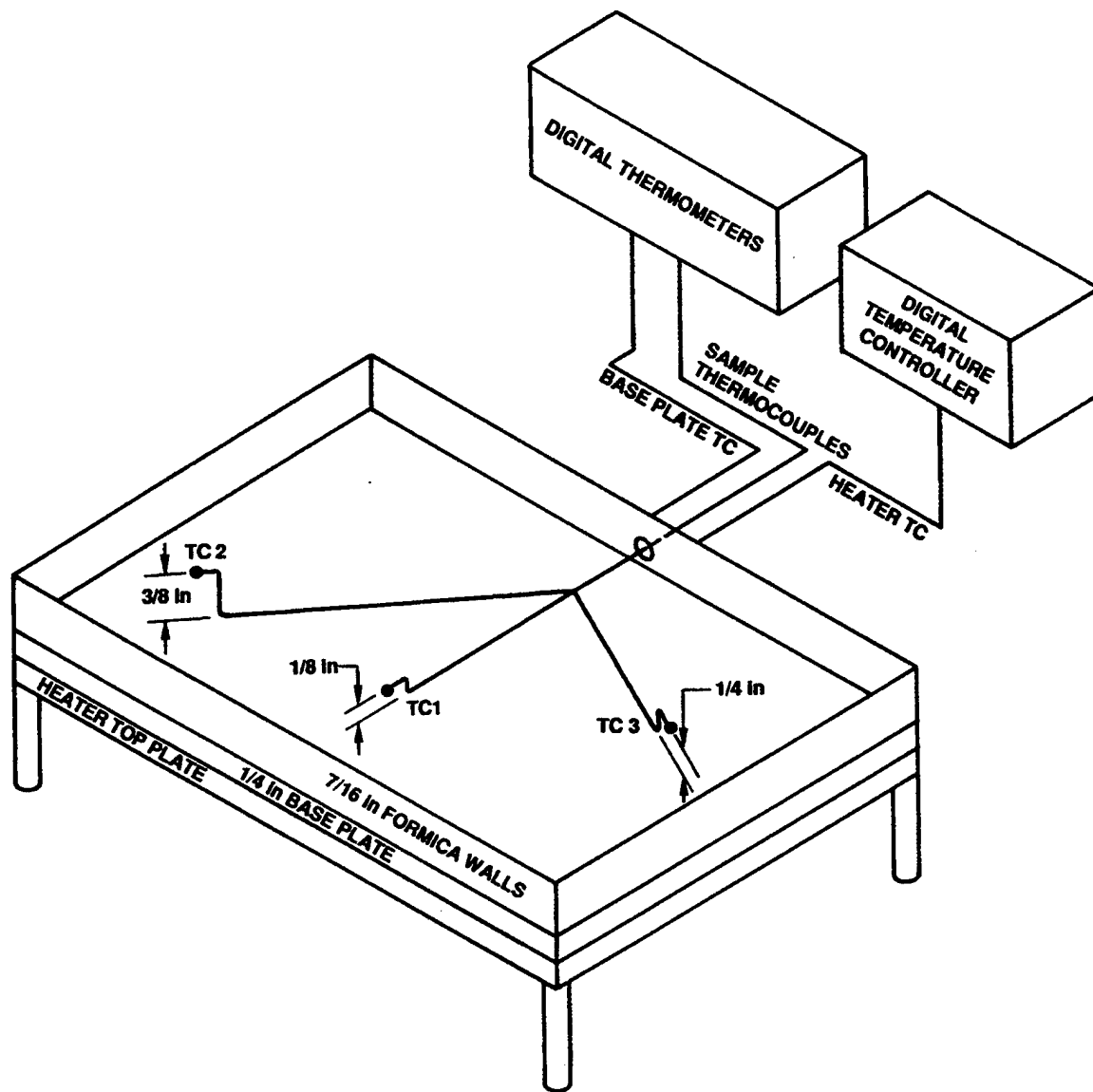


Figure 1. Powder sample heater. (The sample powders were loosely packed into this heater for measurements with the Network Analyzer. The surfaces were smoothed off parallel to the bottom plate prior to measurements.)

measured the temperature during the runs. The reported temperature is an average of temperatures measured at the three different depths within the sample (Cornelison et al. 1992). Temperature variations within the samples are generally on the order of  $10^{\circ}$  C or less.

An almost fully dense (>95% density) potassium nitrate sample was produced by pressing potassium nitrate powder (to 11 ksi) in a 15.2-cm-square mold to a thickness of 8.1 mm. This sample was attached to an aluminum backing plate with conductive paste and then placed on top of the top plate of the heater. During the heating process, another aluminum plate with connected thermocouples was placed on top of the potassium nitrate sample. When the temperatures of the heater plate and the top plate were within 5° C of each other, the top was quickly removed and the measurement taken. Because of this, the temperature measurement of the pressed powder sample was more accurate than for the loose powder samples.

The radar reflection measurements were taken from 2 to 20 GHz using an HP8510B-based centimeter-wave reflectometer (Cornelison et al. 1992). This reflectometer is in the standard form of an arch with the sample resting on a pedestal surrounded by radar-absorbing material. The bistatic angle for these tests was 5°. During each test, the empty sample holder was first measured to obtain a baseline measurement, and then the sample was added for the temperature sweeps. The samples were measured through several temperature cycles, and the data were found to be quite reproducible.

### 3. THEORY

The centimeter-wave dielectric properties of sodium and potassium nitrates and nitrites are discussed in this section. The dielectric losses experienced in these materials are primarily the result of two fundamental loss mechanisms, one involving permanent dipole moments and the other involving dynamically induced dipole moments.

In sodium and potassium nitrite, the charges making up the nitrite ions are geometrically arranged in such a way as to produce a permanent dipole moment (Megaw 1973). At 300 K and above, it is believed that the nitrite ions in potassium nitrite are dynamically disordered and undergo quasi-free rotation. An external field will couple directly to the dipole moment, drive the rotations, and cause a weak, temperature-dependent dielectric loss (Adams, Pogson, and Sharma 1988). For sodium nitrite, the nitrite ions are ordered below 440 K and show little microwave activity (Megaw 1973). As the temperature rises toward 440 K, the nitrite ions initiate a 180° hopping motion and thus begin to couple to the microwave field. As the temperature is raised to 440 K and above, the nitrite ions go collectively into quasi-free rotations and produce a dielectric response qualitatively similar to potassium nitrite (Lu, Qi, and Hardy 1993).

The nitrates show a more subtle behavior than the nitrites. The nitrate ion has no permanent dipole moment since its charges lie on the vertices of an equilateral triangle (Megaw 1973). Thus, the nitrate ions in sodium nitrate do not, to lowest order, couple to a microwave field when they begin to rotate. Also, the sodium ions lie exactly between the centroids of the nitrate ion triangles and therefore are unaffected by the nitrate ion rotations.

In potassium nitrate, the situation is more complex. From room temperature up to  $\sim 128^\circ\text{C}$ , it exists in an orthorhombic structure known as Phase II. If heated to above  $128^\circ\text{C}$ , it undergoes a order/disorder transition to a high-temperature dynamically disordered trigonal structure designated as Phase I. This transition is accompanied by a large expansion along the  $\vec{c}$  axis. Upon cooling from the high-temperature phase, potassium nitrate undergoes a transition into a metastable ferroelectric phase, Phase III. This phase exists from  $\sim 124^\circ\text{C}$  to  $\sim 110^\circ\text{C}$ , below which a transition back to Phase II takes place.

Figure 2 shows two orthogonal projections of a room temperature, Phase II representation of the potassium nitrate lattice. When potassium nitrate is in this stable phase, the structure is orthorhombic Pmcn (Megaw 1973; Lu and Hardy 1991) with the  $\text{K}^+$  ions lying between the planar  $\text{NO}_3^-$  ions along the  $\vec{c}$  axis, but displaced along the  $\vec{b}$  axis so they do not lie above or below the centroids of the  $\text{NO}_3^-$  ions, as shown. In this phase, there is no net static dipole moment. That this phase is nonpolar results from the fact that although there are dipoles associated with the  $\text{K}-\text{NO}_3^-$  chains along the  $\vec{c}$  axis, these occur in antiparallel pairs producing zero net moment. However, when the temperature rises, the  $\text{NO}_3^-$  ions are thermally activated into hindered rotation, and polar fluctuations occur in each chain. These do not cancel in pairs being, at best, only loosely correlated. It is these polar fluctuations that cause the dielectric losses that are observed up to about  $128^\circ\text{C}$ . If the system is heated to above  $128^\circ\text{C}$  into the high-temperature phase, Phase I, there is a radical restructuring involving: (1) the disordering of the  $\text{NO}_3^-$  ions by hindered, almost free rotation about the  $\vec{c}$  axis, and (2) a large displacement of the  $\text{K}^+$  ions within the  $\vec{ab}$  plane such that they move to a position immediately below the centroids of the  $\text{NO}_3^-$  ions, thus causing the large  $\vec{c}$  axis expansion of  $\sim 5\%$ . In this high-temperature phase, since the  $\text{NO}_3^-$  ions are in almost free rotation, they thus produce six-fold rather than three-fold rotational symmetry about the  $\vec{c}$  axis. They also then move to the midpoint of the adjacent  $\text{K}^+$  planes. This motion should cause the polarity to vanish. However, this is only true on average, and about this average configuration there will be continuing fluctuations. This motion is polar and will couple to an external electric field; hence, the observed dielectric absorption in Phase I.

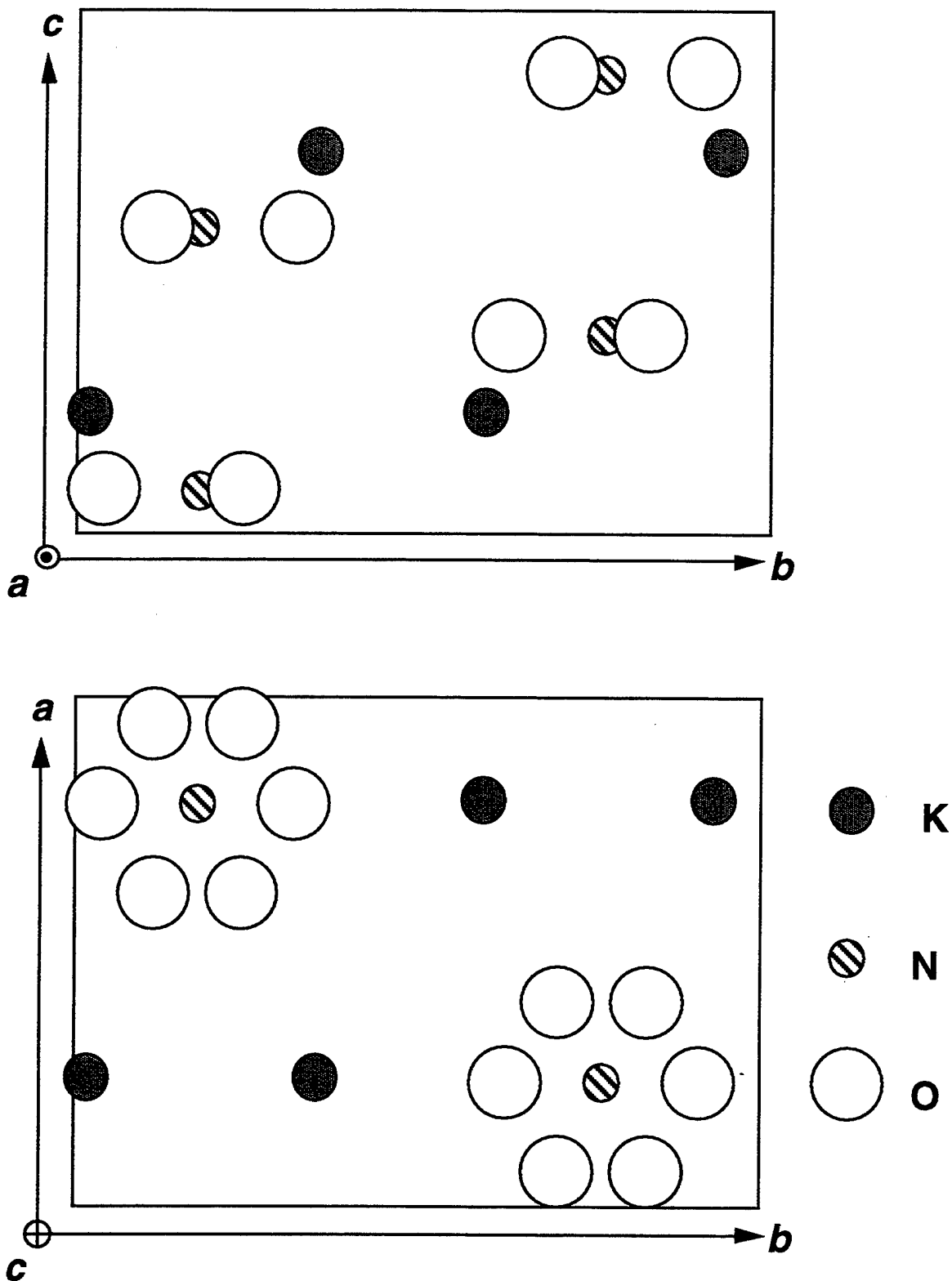


Figure 2. Phase II representation of potassium nitrate lattice. (The top figure shows projections of potassium nitrate ions onto the  $\vec{cb}$  plane. Note the position of the  $K^+$  ions above or below the planes of potassium nitrate. The bottom figure shows the potassium nitrate lattice looking along the  $\vec{c}$  axis.)

If now the crystal is cooled below  $\sim 128^\circ\text{C}$ , it transforms to a new phase, Phase III. In this ferroelectric phase, the  $\text{NO}_3^-$  ions are now aligned so that they are all superposed when viewed down the  $\vec{c}$  axis, but the  $\text{K}^+$  ions do not lie halfway between them, being displaced from the midpoint by a finite distance. The  $\text{K}^+$  ions remain located immediately above or below the  $\text{NO}_3^-$  ion centroids. This causes the development of a spontaneous dipole moment along  $\vec{c}$ , which may be switched by reversing the displacement of the  $\text{K}^+$  ions to the other energetically equivalent structure. This implies a "double well" potential energy surface for the lattice, qualitatively similar to that in barium titanate ( $\text{BaTiO}_3$ ) (Cohen 1992; Cohen and Krakauer 1990, 1992). In both the ferroelectric phase and in Phase II (room temperature, stable), the basic impediment to full but hindered rotation of the  $\text{NO}_3^-$  molecular ions is the presence of the layers of  $\text{K}^+$  ions.

#### 4. RESULTS AND DISCUSSION

Reflection measurements for "50% dense powder" sodium nitrate at frequencies from 2 to 20 GHz and at temperatures up to  $240^\circ\text{C}$  are shown in Figure 3. The losses are small even as the sample is heated above the order/disorder transition temperature. This is expected because of the high symmetry of this system and lack of a dynamically induced dipolar loss mechanism. These results are similar to those obtained for the ionic salts potassium bromide and sodium chloride (Cornelison et al. 1992).

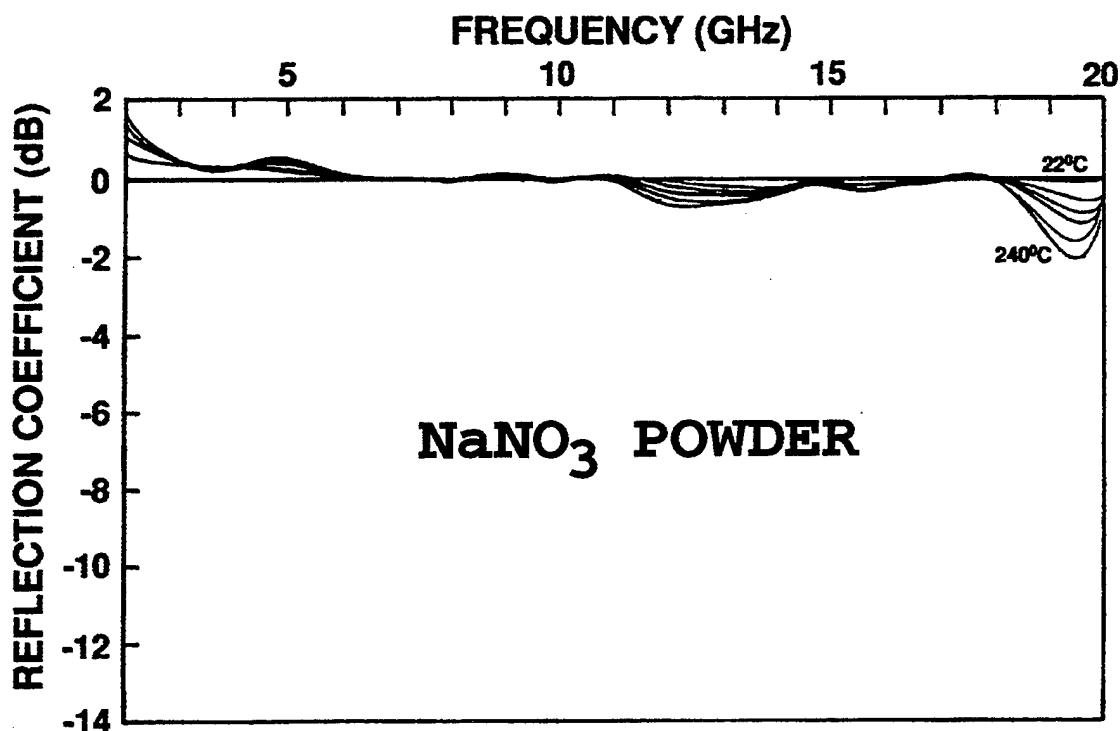


Figure 3. Temperature-dependent microwave reflection curves for sodium nitrate powder sample.

Figures 4–7 show reflectometer data and theoretical fits based on a Debye relaxation model for potassium nitrite, sodium nitrite, and potassium nitrate. The experimental data for potassium and sodium nitrite are for 50% density powders that are more than 1 cm thick. The potassium nitrate results were obtained from an almost fully dense compacted powder sample with a thickness of 8.1 mm. The theoretical fits were obtained from a Debye form (Kittel 1953) for the complex dielectric constant as follows:

$$\epsilon = \epsilon_{\infty} + \frac{4\pi a}{1+i2\pi\nu\tau} = \epsilon' + i\epsilon'',$$

and

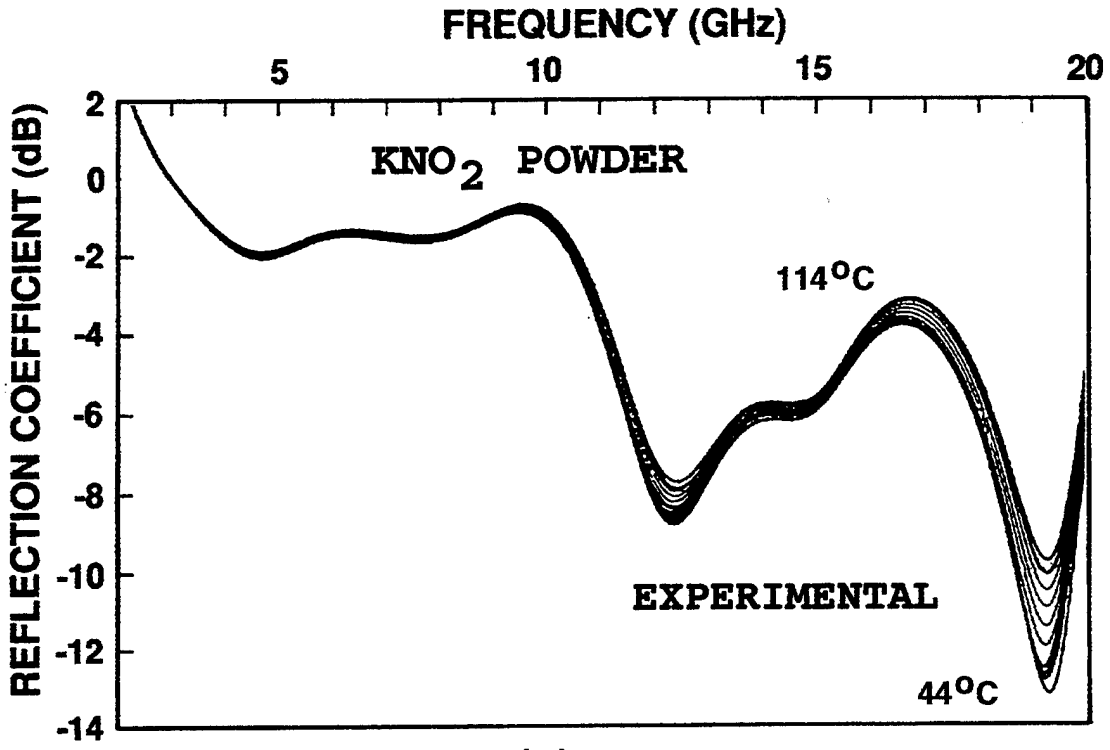
$$\epsilon' = \epsilon_{\infty} + \frac{4\pi a}{1+4\pi^2\nu^2\tau^2} \quad \text{and} \quad \epsilon'' = \frac{8\pi^2 a\nu\tau}{1+4\pi^2\nu^2\tau^2},$$

where

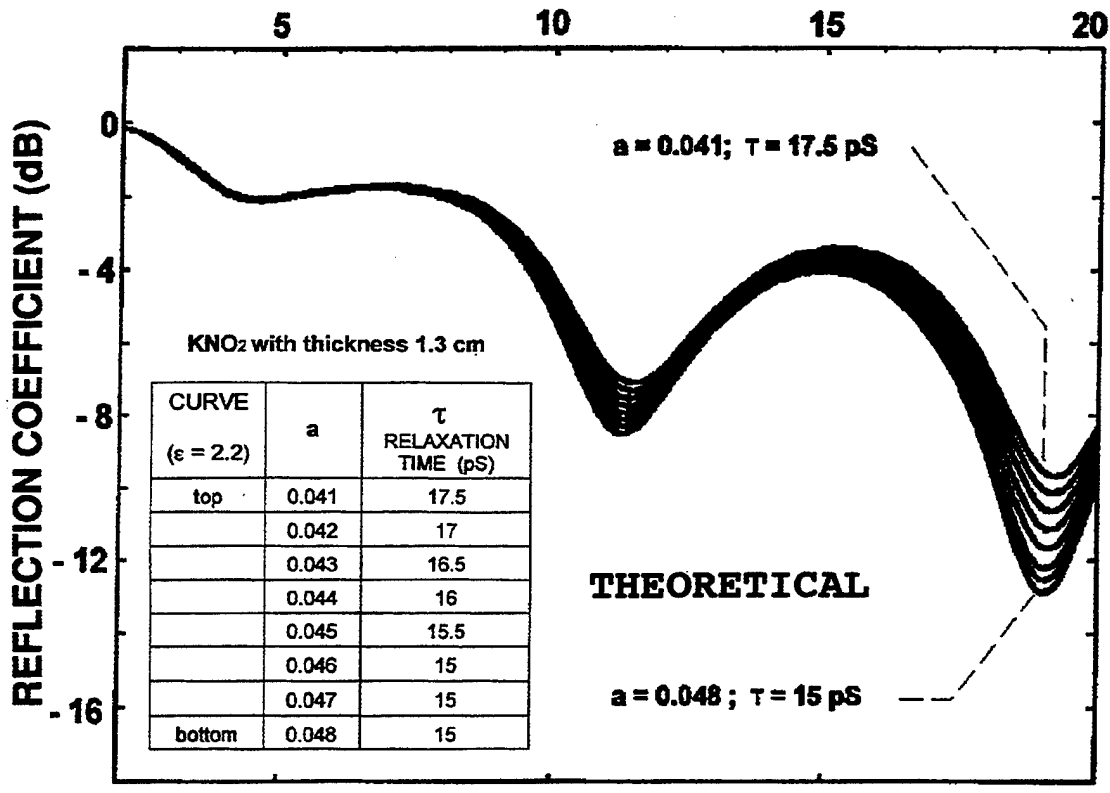
- $\epsilon_{\infty}$  = high-frequency dielectric constant
- $a$  = temperature-dependent parameter
- $\nu$  = frequency of incident radiation
- $\tau$  = relaxation time,

and where  $\tau =$  and  $a =$  were adjusted to produce the best overall fit to the experimental absorption/reflectivity data.

Figure 4(a) shows the results of measurements on a 1.3-cm-thick, loosely packed powder of potassium nitrite at temperatures from 46° C to 114° C. Significant losses occur above room temperature through 114° C with a weak temperature variation. This Debye-like temperature dependence is in agreement with what is expected for this type of a polar solid (Kittel 1971; Blakemore 1969). Figure 4(b) shows the theoretical fits to the experimentally obtained temperature data for potassium nitrite. The parameters chosen for the fits are shown in the table contained within Figure 4(b). The value of  $\epsilon_{\infty} = 2.2$  was chosen as a reasonable value for less than full density loosely packed powder of potassium nitrite. Although some smaller features of the experimental data are not reproduced in the fits, generally the fits are quite good with the chosen parameters.

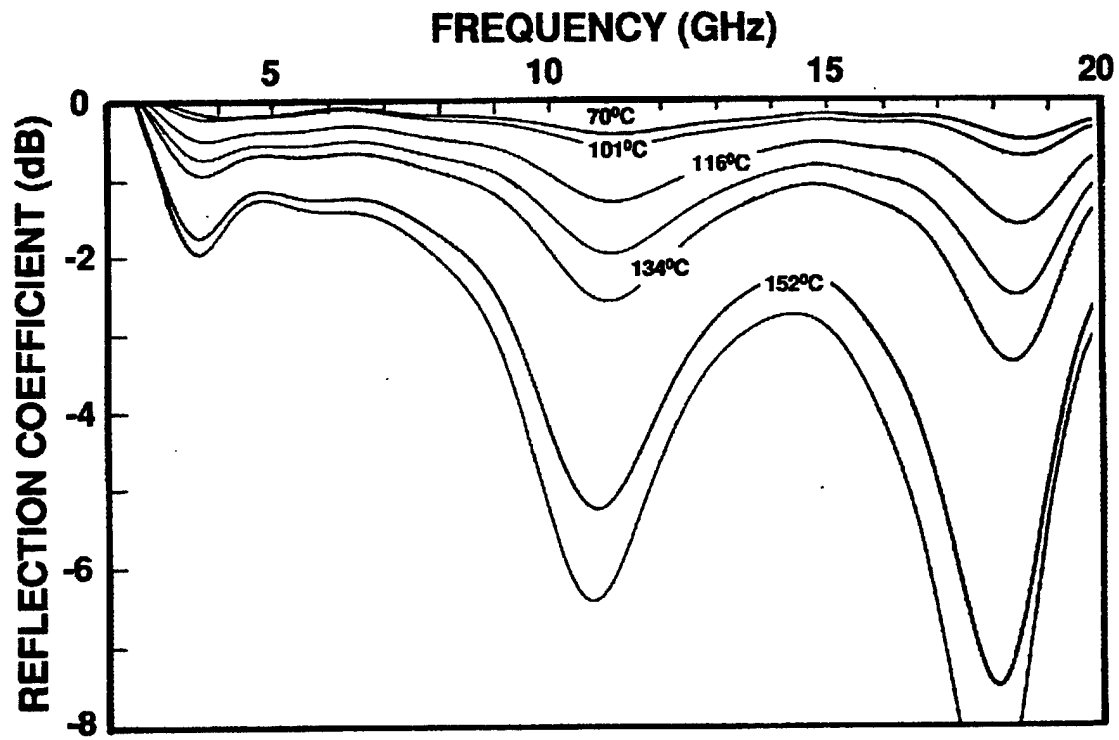


(A)

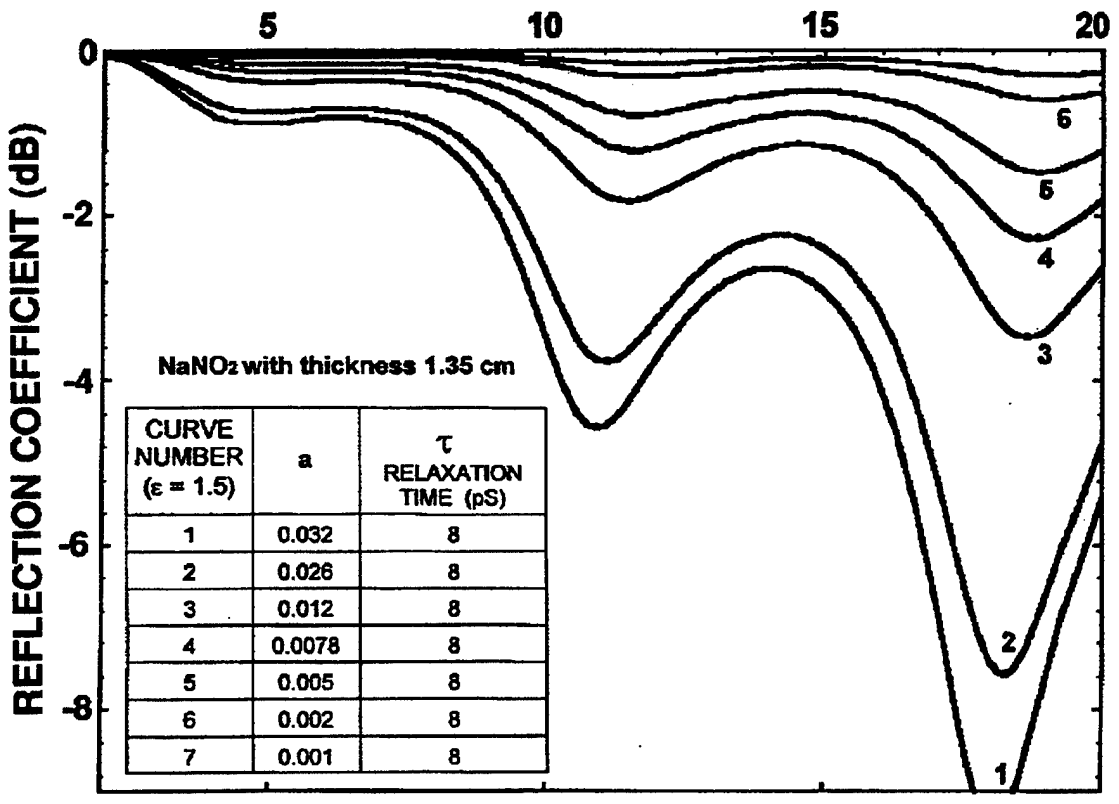


(B)

Figure 4. Experimental reflectometer measurements and theoretical calculations for potassium nitrite at various temperatures.

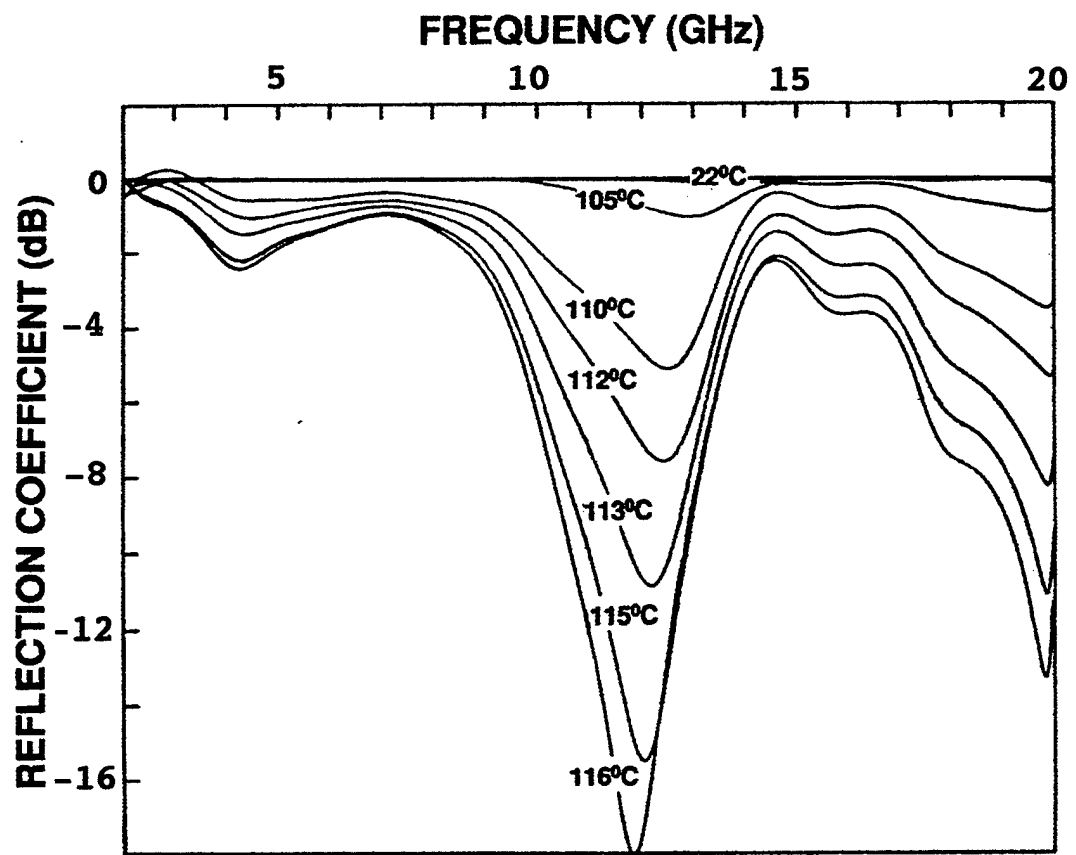


(A)

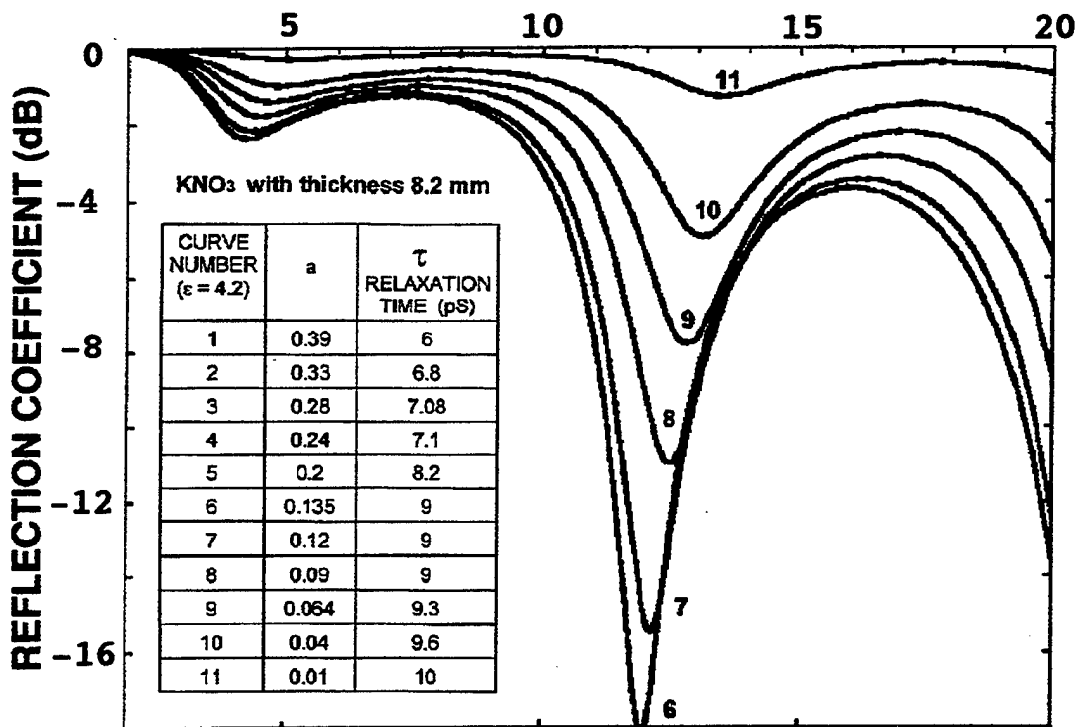


(B)

Figure 5. Experimental reflectometer measurements and theoretical calculations for sodium nitrite at various temperatures.

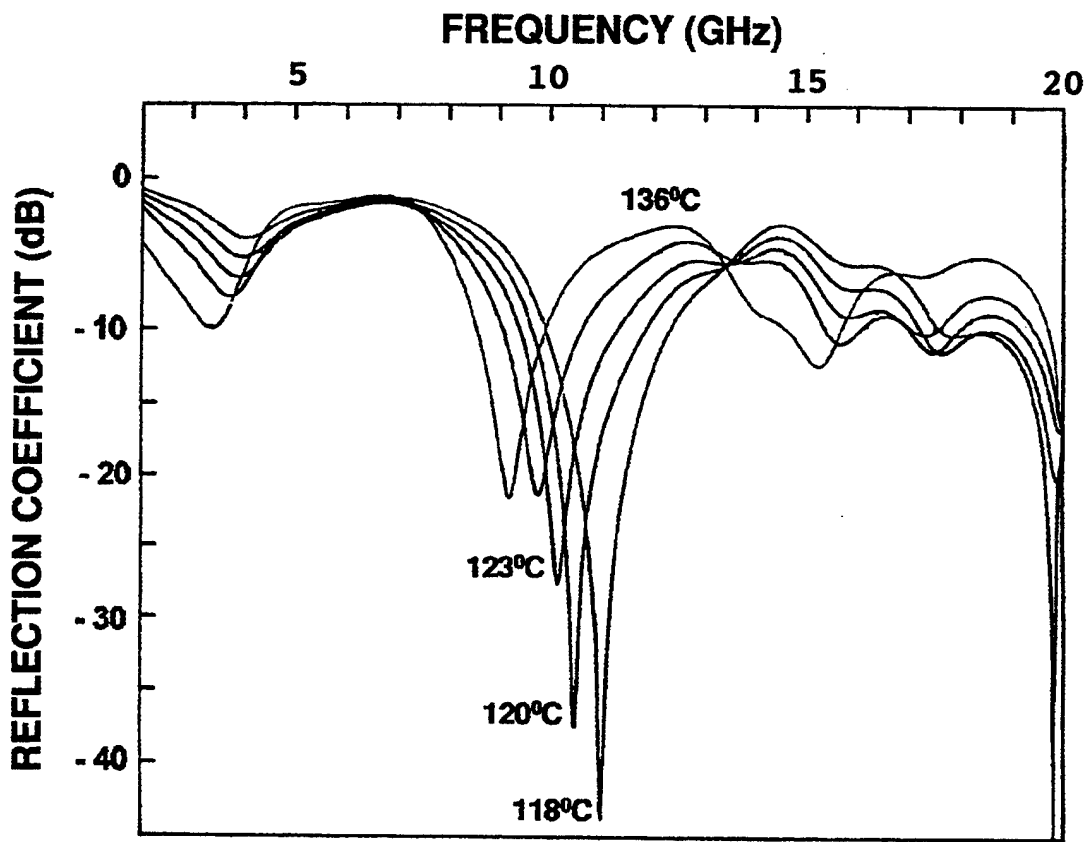


(A)

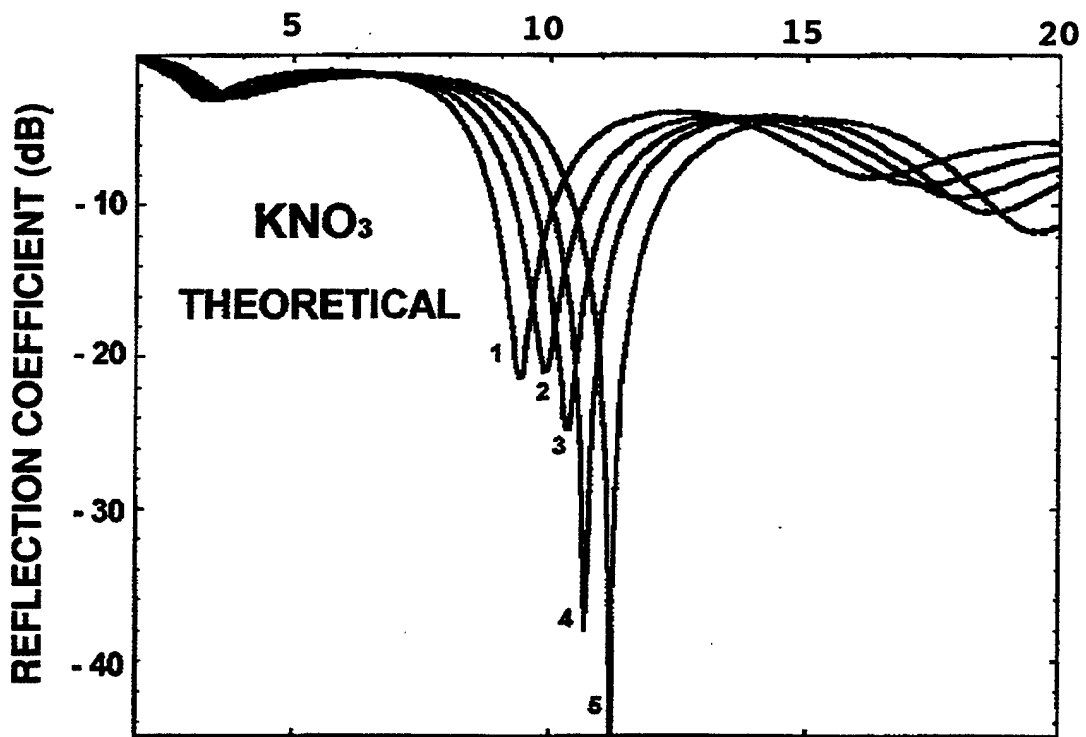


(B)

Figure 6. Experimental reflectometer measurements and theoretical results from room temperature to 116° C for almost fully dense pressed powder potassium nitrate.



(A)



(B)

Figure 7. Experimental reflectometer measurements and theoretical results from 118° C to 136° C for almost fully dense pressed powder potassium nitrate.

Figure 5(a) shows experimental data for sodium nitrite from room temperature to above 150° C. Experimentally, below 70° C, there is little change from the baseline measurements. Above 100° C, the losses begin to develop and increase dramatically up to above 150° C. This implies that some thermally activated hopping of the nitrite ions occurs below the transition temperature of 440 K (167° C). Figure 5(b) shows the theoretical fits to the sodium nitrite data with the parameters as indicated within the figure. Although the strengths of some of the reflection minima are not perfectly represented in the theoretical fits, the fits obtained with a single parameter ( $a = 0.032$  to  $0.001$ ) represent the data fairly well over the temperature range measured.

Results of experimental measurements and theoretical fits on a nearly fully dense, 8.1-mm-thick, pressed powder sample of potassium nitrate are shown in Figures 6 and 7. The temperature was raised from room temperature to about 136° C. No attempt was made to measure the sample in the ferroelectric phase (Phase III) upon cooling from the high-temperature phase. In Figure 6(a), experimental data are shown from room temperature to 116° C. In Figure 6(b), corresponding theoretical fits are shown. The table contained in Figure 6(b) shows the parameters utilized to obtain the fits for both Figures 6 and 7. In Figure 7(a), experimental data from 118° C to 136° C are shown with the corresponding theoretical fits shown in Figure 7(b). As can be seen in the figures, reflection losses begin to occur as the temperature is raised to and above 100° C. Above this temperature, the two large reflection minima move to lower frequencies and become weaker. A smaller, low-frequency minimum (below 5 GHz) continues to grow as the temperature is raised to above 136° C. The results on potassium nitrate are strong evidence for the development of a dipolar loss mechanism above ~100° C as discussed previously. The weakening of the losses above the transition can be explained by the movement of the potassium ions toward the centroid line joining the nitrate ions. This results in a decrease in the dynamically induced dipole moment and thus the mechanism which leads to loss in this material.

When one compares theory with experiment, it is evident that the agreement with experiment is remarkably close given that only two parameters are employed. Indeed, one can obtain fits almost as good by varying the parameter "a" alone. However, the modest variations in  $\tau$  that have been employed are consistent with the  $(T)^{-1}$  dependence that one expects from high-temperature phonon damping.

More germane to the theoretical fits to experiment is the derivation of the values of "a" from basic principles. The simplest possible approach is to assume that these losses are due to arrays of *independent*

dipoles whose moments can be derived or inferred from the structural data. The resultant expression for the coefficient "a" is:

$$a = \frac{N\eta^2}{KT},$$

where N is the number of dipoles/unit volume,  $\eta$  is the magnitude of this moment, K is Boltzmann's constant, and T is the temperature (Kittel 1963).

For the potassium nitrate dipole, we can approximate  $\eta$  by the polarization per unit cell in the metastable ferroelectric phase (Phase III) obtained from the observed and theoretical values of  $\approx 10 \frac{\mu\text{C}}{\text{cm}^2}$  (Chen and Chernow 1967).

If we apply the previously given values, we find that  $a \approx 1.5$ , giving  $\epsilon(\omega \rightarrow 0) \approx 20$  for a field parallel to the polar axis of Phase I with the use of  $\epsilon_\infty = 4.2$ . However, Chen and Chernow (1967) report a value for  $\epsilon(\omega \rightarrow 0) \approx 50-60$  for Phase I, when the electric field is applied along the  $\vec{c}$  axis. This difference is not a serious problem since our independent dipole model neglects two significant effects, both of which will enhance  $\epsilon \parallel \vec{c}$ : (1) any correlation between dipoles (e.g., a mean field term), and (2) allowance for local field effects.

The major problem is that, since our specimens are pressed powders, and we only expect  $\epsilon \parallel \vec{c}$  to show these much enhanced values, then our experimental value of  $\epsilon(\omega \rightarrow 0)$  should be  $\sim \frac{1}{3} \epsilon(\omega \rightarrow 0) \parallel \vec{c}$  *i.e.*  $\approx 20$ . This requires  $a \approx 1.5$ , which is almost triple the largest value obtained from the fits to the experimental data.

Clearly, we have a major discrepancy, and the most likely explanation is nonuniformity of temperature across the specimens, which are approximately 1 cm thick. Since the transition is very sharp and jumps discontinuously from  $\approx 10$  to  $\approx 50-60$  at transition, it appears likely there will be a sharp interface between the transformed and the untransformed material that will travel from the metal backing to the surface as the former is heated. Thus, the absorption that is actually measured is loss averaged over the specimen

thickness, with most of the loss probably coming from a thin layer whose temperature is at or very close to  $T_c$ . Thus, the effective thickness is much less than the actual width.

In the case of the nitrites, sodium nitrite shows analogous behavior to potassium nitrate except that the nature of the dipoles is different as we have discussed previously. However, it appears that none of the specimen has actually undergone transformation into the high-temperature phase.

## 5. SUMMARY

We have studied the temperature-dependent centimeter-wave reflection properties of powdered samples of several molecular crystals. The results of reflectometer temperature-dependent measurements were compared to theoretical calculations based on a Debye relaxation model for the potassium nitrate, potassium nitrite, and for the sodium nitrite samples. The nitrite samples showed absorptions when temperatures were high enough to excite "hopping" modes. For potassium nitrite, losses were seen at all of the measured temperatures, and sodium nitrite showed losses above  $\sim 100^\circ\text{C}$ , below the  $165^\circ\text{C}$  transition temperature. Sodium nitrate showed little losses, but potassium nitrate showed interesting behavior around or above the order/disorder transition temperature of  $\sim 128^\circ\text{C}$ . This behavior is very likely caused by a dynamically induced dipolar loss mechanism.

## 6. REFERENCES

- Adams, D. M., M. Pogson, and S. K. Sharma. "Spectroscopy at Very High Pressures: Part 51. A Raman and Infrared Study of the Polymorphs of  $\text{KNO}_2$ ." Journal Phys. Chem: Solid State Phys., vol. 21, 1988.
- Blakemore, J. S. Solid State Physics. Philadelphia, PA: W. B. Saunders, 1969.
- Chen, A., and F. Chernow. "Nature of Ferroelectricity in  $\text{KNO}_3$ ." Physical Review, vol. 154, no. 2, pp. 493-505, 1967.
- Cohen, R. E. Nature, vol. 358, p. 136, 1992.
- Cohen, R. E., and H. Krakauer. Physical Review B, vol. 42, p. 6416, 1990.
- Cohen, R. E., and H. Krakauer. Ferroelectrics, vol. 136, p. 65, 1992.
- Cornelison, S. G., A. Gauss, Jr., R. B. Bossoli, H. M. Lu, and J. R. Hardy. "Centimeter Wave Absorption in Molecular Crystals." BRL-MR-4002, U.S. Army Ballistic Research Laboratory, Aberdeen Proving Ground, MD, September 1992.
- Jona, F., and G. Shirane. Ferroelectric Crystals. New York: The Macmillan Company, 1962.
- Kittel, C. Introduction to Solid State Physics, first edition, New York: J. Wiley, p. 89, 1953.
- Kittel, C. Introduction to Solid State Physics, second edition, London: J. Wiley and Sons, Inc., 1963.
- Kittel, C. Introduction to Solid State Physics, fourth edition, New York: J. Wiley and Sons, 1971.
- Lu, H. M., and J. R. Hardy. "Ab Initio Simulations of Phase Transitions in  $\text{KNO}_3$ ." Ferroelectrics, vol. 111, pp. 43-47, 1990.
- Lu, H. M., and J. R. Hardy. "First Principles Study of Phase Transitions in  $\text{KNO}_3$ ." Physical Review B, vol. 44, no. 14, October 1991.
- Lu, H. M., R. Qi, and J. R. Hardy. "A Prior Study of the Order-Disorder Phase Transition in  $\text{NaNO}_2$ ." Solid State Communications, vol. 87, no. 12, pp. 1151-1154, 1993.
- Megaw, H. D. Crystal Structures, A Working Approach. Philadelphia, PA: B. Saunders, 1973.
- Von Hippel, A. R. Dielectric Materials and Applications. Massachusetts Institute of Technology Press, 1954.

INTENTIONALLY LEFT BLANK.

NO. OF  
COPIES      ORGANIZATION

2      DEFENSE TECHNICAL INFO CTR  
ATTN DTIC DDA  
8725 JOHN J KINGMAN RD  
STE 0944  
FT BELVOIR VA 22060-6218

1      DIRECTOR  
US ARMY RESEARCH LAB  
ATTN AMSRL CS AL TP  
2800 POWDER MILL RD  
ADELPHI MD 20783-1145

1      DIRECTOR  
US ARMY RESEARCH LAB  
ATTN AMSRL CS AL TA  
2800 POWDER MILL RD  
ADELPHI MD 20783-1145

3      DIRECTOR  
US ARMY RESEARCH LAB  
ATTN AMSRL CI LL  
2800 POWDER MILL RD  
ADELPHI MD 20783-1145

ABERDEEN PROVING GROUND

2      DIR USARL  
ATTN AMSRL CI LP (305)

NO. OF  
COPIES

ORGANIZATION

- 3 DIRECTOR  
US ARMY RESEARCH LAB  
ATTN AMSRL LT  
ROBERT WEINRAUB  
DON WOODBURY  
MICHAEL MATTIE  
2800 POWDER MILL RD  
ADELPHI MD 20783-1145
- 2 DIRECTOR  
US ARMY RSRCH OFC  
ATTN M CLIFTON  
PO BOX 12211  
RESEARCH TRIANGLE PARK NC  
27709-221
- 12 UNIV OF NEBRASKA AT LINCOLN  
DEPT OF PHYSICS AND  
CTR FOR ELECTRO OPTICS  
ATTN J HARDY 6 CPS  
J KRANE 6 CPS  
LINCOLN NE 68504

ABERDEEN PROVING GROUND

- 26 DIR, USARL  
ATTN: AMSRL-WT-W, C MURPHY  
AMSRL-WT-WD,  
S. CORNELISON (25 CP)

USER EVALUATION SHEET/CHANGE OF ADDRESS

This Laboratory undertakes a continuing effort to improve the quality of the reports it publishes. Your comments/answers to the items/questions below will aid us in our efforts.

1. ARL Report Number/Author ARL-TR-1179 (Cornelison) Date of Report August 1996

2. Date Report Received \_\_\_\_\_

3. Does this report satisfy a need? (Comment on purpose, related project, or other area of interest for which the report will be used.) \_\_\_\_\_

4. Specifically, how is the report being used? (Information source, design data, procedure, source of ideas, etc.) \_\_\_\_\_

5. Has the information in this report led to any quantitative savings as far as man-hours or dollars saved, operating costs avoided, or efficiencies achieved, etc? If so, please elaborate. \_\_\_\_\_

6. General Comments. What do you think should be changed to improve future reports? (Indicate changes to organization, technical content, format, etc.) \_\_\_\_\_

CURRENT ADDRESS

Organization

Name

Street or P.O. Box No.

City, State, Zip Code

7. If indicating a Change of Address or Address Correction, please provide the Current or Correct address above and the Old or Incorrect address below.

OLD ADDRESS

Organization

Name

Street or P.O. Box No.

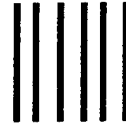
City, State, Zip Code

(Remove this sheet, fold as indicated, tape closed, and mail.)  
(DO NOT STAPLE)

---

**DEPARTMENT OF THE ARMY**

OFFICIAL BUSINESS



**NO POSTAGE  
NECESSARY  
IF MAILED  
IN THE  
UNITED STATES**

**BUSINESS REPLY MAIL**  
FIRST CLASS PERMIT NO 0001,APG,MD

POSTAGE WILL BE PAID BY ADDRESSEE

**DIRECTOR  
U.S. ARMY RESEARCH LABORATORY  
ATTN: AMSRL-WT-WD  
ABERDEEN PROVING GROUND, MD 21005-5066**

