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**RESEARCH ON FERROELECTRIC MATERIALS
FOR MILLIMETER WAVE APPLICATION**

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62-10814-206 (SBN:60). All systems examined to date exhibit high dielectric loss at millimeter wavelengths, the minimum being $\tan \delta = 0.3$ near 40 GHz for a high purity SBN:60 sample. The generality of this observation, when coupled with high sample-to-sample variability, suggests a commonly occurring extrinsic factor, such as growth defects, as the key element. How the frequency scale for polarization fluctuations is tied to such extrinsic influences remains to be explored. A systematic study of the influence of defect structure on microwave dielectric properties of ferroelectrics is probably the most appropriate next step.

*Sr.6Ba.4Nb2O6

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TABLE OF CONTENTS

	<u>Page</u>
1.0 SUMMARY.....	1
1.1 Technical Problem.....	1
1.2 General Methodology.....	2
1.3 Technical Results.....	2
1.4 Implications for Further Research.....	3
2.0 MATERIALS DEVELOPMENT.....	4
2.1 Introduction.....	4
2.2 Growth of SBN:60 and SBN:50 Single Crystals.....	4
2.3 Tungsten Bronze $Pb_{1-2x}K_xM_x^{3+}Nb_2O_6$	7
2.4 Perovskite $Ba_{1-x}Sr_xTiO_3$ System.....	8
2.5 Future Materials.....	14
3.0 HIGH FREQUENCY DIELECTRIC MEASUREMENTS.....	15
3.1 SBN:60 Crystals.....	15
3.2 Lead Potassium Lanthanum Niobate (PKLN) System.....	18
3.3 Barium Strontium Titanate (BST) System.....	18
4.0 FUTURE PLANS.....	19
5.0 PUBLICATIONS AND PRESENTATIONS.....	20
5.1 Publications.....	20
5.2 Presentations.....	20
6.0 REFERENCES.....	21

LIST OF TABLES

<u>Table</u>	<u>Page</u>
1 Physical Properties of Various Ferroelectric Materials.....	5

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LIST OF FIGURES

<u>Figure</u>		<u>Page</u>
1	Phase boundary and Curie temperatures vs composition for $Sr_{1-x}Ba_xNb_2O_6$	6
2	Variation of ferroelectric transition temperature for $Pb_{1-2x}K_xM_x^{3+}Nb_2O_6$, M = La or Bi.....	9
3	Phase diagram for ferroelectricity in the solid solution system $Pb_{1-x}Ba_xNb_2O_6$	10
4	Relative permittivity as a function of temperature for $Ba_{.70}Sr_{.30}TiO_3$	12
5	Tan δ as a function of temperature for $Ba_{.70}Sr_{.30}TiO_3$	13
6	Temperature dependence of imaginary relative permittivity at 30, 33, and 36 GHz.....	16
7	Temperature dependence of real relative permittivity at 30, 33, and 36 GHz.....	17



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1.0 SUMMARY

This technical report presents the results of the second six months of a research program on ferroelectric materials for phase control applications in millimeter wave radar systems. Originally a one-year program, this effort has been extended to fifteen months to take advantage of materials development activities in related programs. The major accomplishments in the current period include growth and preparation of several ferroelectric materials, and high frequency dielectric characterization of high purity single crystals of $\text{Sr}_{.6}\text{Ba}_{.4}\text{Nb}_2\text{O}_6$ (SBN:60).

1.1 Technical Problem

On the basis of current models for ferroelectric materials, one predicts that certain ferroelectrics having a high dc permittivity $\epsilon(0)$ should also show high sensitivity of their microwave refractive index, $n = \sqrt{\epsilon(\omega)}$ to an applied electric field for microwave frequencies up to several hundred GHz. A low absorptive loss is also predicted over the same range of frequencies. However, the millimeter wave dielectric properties of these materials are largely unknown, and growth of the most promising ferroelectrics in single crystal form is generally difficult.

The present program was conceived on the basis of successful growth at Rockwell on one such ferroelectric, $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$, and measurement in this material of a substantial electric field sensitivity, dn/dE , of 10^{-6} meters/volt at 58 GHz.¹ With about an order of magnitude larger sensitivity, one can design microwave components operating at practical control voltages (under 200 volts) for phase shifting, modulation and switching. One attractive concept is a planar dielectric lens for electronically steering a millimeter wave radar beam, which could be used in high speed seeker applications.

The technical objective of the current study is to explore the range of dielectric properties (primarily dn/dE and dielectric loss) achievable within the SBN family. Other promising ferroelectrics are to be examined depending on availability.



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1.2 General Methodology

There is an on-going program in development of growth techniques for tungsten-bronze materials at Rockwell, which provides the principal source for the $Sr_{1-x}Ba_xNb_2O_6$ ferroelectrics to be studied under the present contract. This program has produced the largest and highest quality single crystals reported to date of $Sr_{0.61}Ba_{0.39}Nb_2O_6$, the congruently melting composition.² Having these crystals available as seeds greatly facilitates the Czochralski growth of other SBN compositions and certain other tungsten bronzes. Also, through the ferroelectrics program at Penn State University, a wide variety of other materials has been made available for this study. These include ferroelectric and antiferroelectric ceramics, as well as single crystals, many of which have been well characterized by piezoelectric and low frequency dielectric measurement techniques.

Accurate high frequency dielectric measurements on the selected system are being carried out in waveguide from 30 to 100 GHz.³ Power reflection and transmission coefficients are determined on samples cut to fill the guide, and sample dielectric properties are fitted to these observations. The electric field sensitivity of the microwave refractive index, dn/dE , is to be evaluated by a phase bridge technique in the same waveguide geometry.

1.3 Technical Results

During the first six months of this program, millimeter wave measurements on SBN:60 single crystals revealed unexpected behavior which does not fit accepted models for the dielectric properties. Principally, a high level of dielectric loss was found in all samples, and relatively large decreases were noted in permittivity from the low frequency values. Since the envisioned phase control applications require much lower loss, the source of the observed high loss and its dependence on controllable factors has become our primary interest.

In the current period, several SBN:60 single crystal samples grown from ultra-pure chemicals have been characterized from 30 to 40 GHz, and from

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90 to 100 GHz. Measurements of polar axis dielectric properties, both above and below the Curie point, were made from 30-40 GHz. In addition, measurements on two ceramic systems, lead potassium lanthanum niobate (PKLN) and barium strontium titanate (BST), have been undertaken. These systems possess unusual low frequency dielectric properties which may provide viable alternatives for achieving phase control.

1.4 Implications for Further Research

All systems examined to date exhibit high dielectric loss at millimeter wavelengths, the minimum being $\tan \delta_{11} \sim 0.3$ near 40 GHz for a high purity SBN:60 sample. The generality of this observation, when coupled with high sample-to-sample variability, suggests a commonly occurring extrinsic factor, such as growth defects, as the key element. How the frequency scale for polarization fluctuations is tied to such extrinsic influences remains to be explored. A systematic study of the influence of defect structure on microwave dielectric properties of ferroelectrics is probably the most appropriate next step.



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2.0 MATERIALS DEVELOPMENT

2.1 Introduction

Because of the previously reported high losses ($\tan\delta > 0.1$) at millimeter wave frequencies for the tungsten-bronze compositions, $\text{Sr}_{.6}\text{Ba}_{.4}\text{Nb}_2\text{O}_6$ (SBN:60) and $\text{Sr}_{.5}\text{Ba}_{.5}\text{Nb}_2\text{O}_6$ (SBN:50), during the past six months we have initiated work on other potential candidates for millimeter wave applications. These materials include the orthorhombic bronze systems, $\text{Pb}_{1-2x}\text{K}_x\text{M}_x^{3+}\text{Nb}_2\text{O}_6$, $\text{M} = \text{La}$ or Bi and $\text{Pb}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$, and tetragonal perovskite $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ compositions. The low frequency dielectric properties of these materials are summarized in Table 1, along with data for SBN:60 and SBN:50. The high dielectric constant values for PKLN, PBN and $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ show them to be very promising for millimeter wave applications, and their dielectric properties at millimeter wave frequencies are presently under investigation.

We are continuing our work on the single crystal Czochralski growth of SBN:60 and SBN:50 in an attempt to improve the crystal quality and reduce the dielectric losses for these compositions. The work has focused on the use of high purity starting materials and the installation of a new crystal puller with automatic diameter control. The millimeter wave dielectric properties of these latest SBN crystals are now being evaluated.

2.2 Growth of SBN:60 and SBN:50 Single Crystals

Considerable effort has been made to grow and characterize $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$, $x = 0.6$ and 0.5 , solid solution crystals grown by the Czochralski technique.^{2,4} We have now demonstrated the ability to grow large diameter (2 to 3 cm) SBN:60 and SBN:50 single crystals which show good electrical, mechanical and optical quality for potential use in a wide variety of applications. Although the end members SrNb_2O_6 and BaNb_2O_6 do not belong to the tungsten-bronze structural family, the solid solution $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$ crystallizes in the tetragonal tungsten-bronze structures for $0.25 < x < 0.75$.⁵ The solid solubility range for the three different phases and the variation of



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Table 1
Physical Properties of Various Ferroelectric Materials

Composition	Structure	Curie Temp. °/C	Dielectric Constant	
			Room Temp.	T _c
Sr _{.60} Ba _{.40} Nb ₂ O ₆ *	T.T.B.	72	880	> 50,000
Sr _{.50} Ba _{.50} Nb ₂ O ₆ *	T.T.B.	120		
Pb _{.80} K _{.10} La _{.10} Nb ₂ O ₆	O.T.B.	339	720	3,400
Pb _{.70} L _{.15} La _{.15} Nb ₂ O ₆	O.T.B.	201	790	1,600
Pb _{.70} L _{.15} Bi _{.15} Nb ₂ O ₆	O.T.B.	211	750	2,840
Ba _{.70} Sr _{.30} TiO ₃ (1450°C)	T.P.	20	3000	14,000
Ba _{.70} Sr _{.30} TiO ₃ (1400°C)	T.P.	20	3000	14,000

T.T.B. - Tetragonal tungsten bronze
O.T.B. - Orthorhombic tungsten bronze
T.P. - Tetragonal perovskite
* - Single crystal samples

the ferroelectric phase transition temperature for this solid solution are shown in Fig. 1. The present work on SBN crystals has focused on the x = 0.60 and 0.50 compositions in order to obtain information on the range of properties available within this solid solution system.

Initial measurements on SBN:60 and SBN:50 crystals have shown unexpectedly high dielectric losses ($\tan\delta > 0.1$) in the range of 30-40 GHz and 90 - 100 GHz. Although the source of these losses remain undetermined, if extrinsic factors control their magnitude and frequency dependence, it may be possible to substantially reduce the dielectric losses in the 30-40 and 90-100 GHz range through changes in growth control and preparation. Furthermore, optical striations have been somewhat of a problem in these crystals, indicating that tighter control over growth conditions is necessary for improved sample quality.

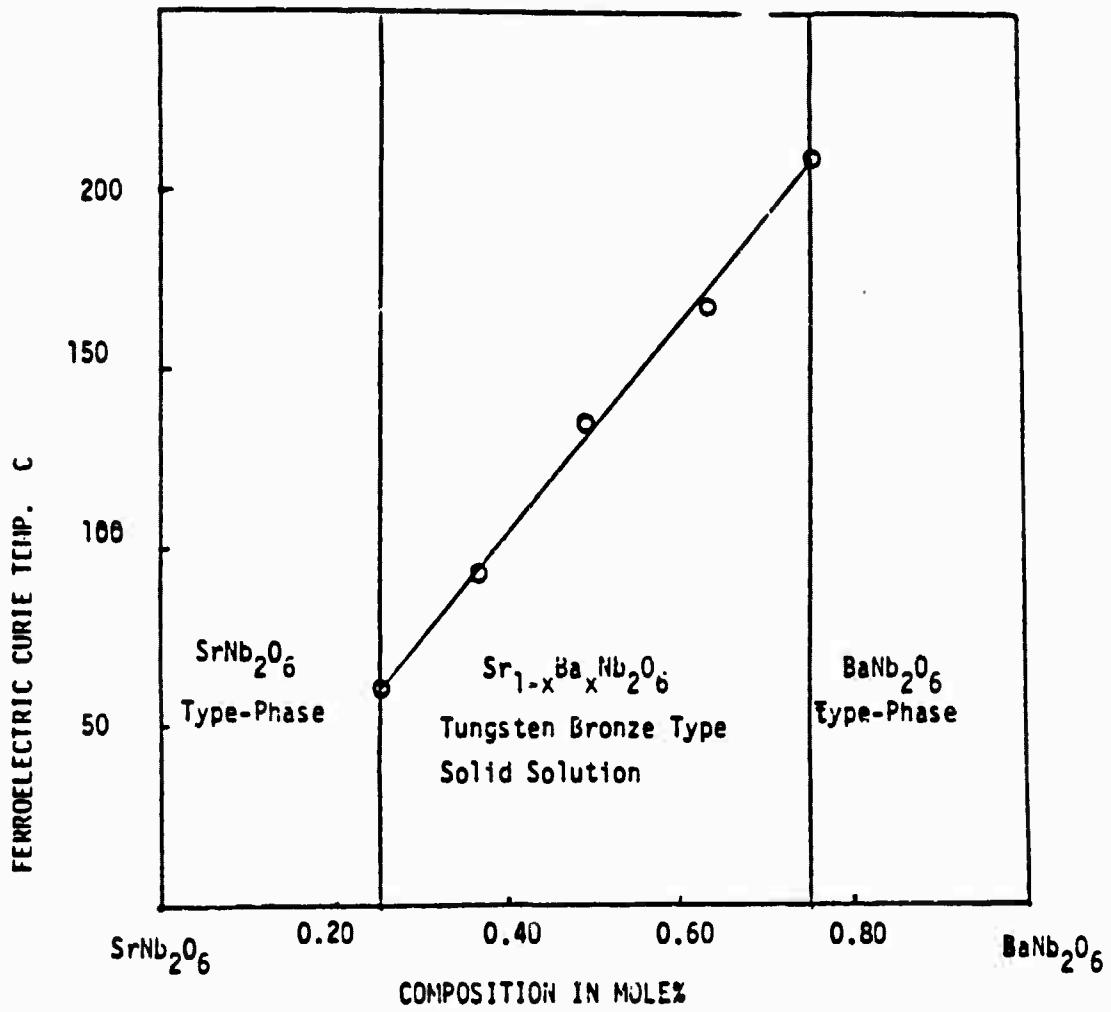


Fig. 1 Phase boundary and Curie temperatures vs composition for $\text{Sr}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$.



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To this end, we have begun SBN crystal growth using high purity starting materials and a new Czochralski puller which features automatic diameter control, improved control over temperature gradients and improved temperature stability. Initial growths of high purity SBN:60 and SBN:50 crystals with this unit show a substantial reduction in striations in comparison with those grown with our earlier unit. Measurements of the millimeter wave dielectric properties of these latest crystals are now in progress.

2.3 Tungsten Bronze $Pb_{1-2x}K_xM_x^{3+}Nb_2O_6$ System

The $Pb_{1-2x}K_xM_x^{3+}Nb_2O_6$ system is based on the orthorhombic tungsten-bronze $PbNb_2O_6$ phase, which was the first oxide-ferroelectric ever discovered that was not a perovskite.^{6,7} $PbNb_2O_6$ has been used commercially as a piezoelectric ceramic transducer. Its outstanding features are its ability to withstand exposure to temperatures approaching its Curie point (570°C) without severe depoling, its large d_{33}/d_{31} ratio, and its extremely low mechanical Q. Above the Curie point, the structure is tetragonal with lattice constants, $a = 12.56$ and $c = 3.925A$, space group $P4/mbm$, isostructural with potassium tungsten-bronze. Below the Curie point, there is slight orthorhombic distortion, quadrupling the cell size, with lattice constants becoming $a = 17.63$, $b = 17.93$ and $c = 7.736A$, space group $Cmm2$.

Although the Curie temperature was much higher than that of any known ferroelectric, the material did not find immediate application because of the difficulty in preparing good nonporous ceramics and the associated problem of poling them. By analogy with previous work on barium titanate and other ferroelectric hosts, the effect of replacing part of Pb^{2+} by other divalent and trivalent ions, or Nb^{5+} by tetravalent or hexavalent ions was studied,⁸⁻¹⁵ with the objective of improving the sintering and general ferroelectric properties of the ceramic. It was observed that the Curie temperature decreased, and although this would be an disadvantage, this made it possible to pole the material more effectively and successfully enhance the dielectric and piezoelectric properties. The systems $Pb_{1-2x}K_xM_x^{3+}Nb_2O_6$, $M = La$ or Bi , and $Pb_{1-x}Ba_xNb_2O_6$ are typical examples of such substitutions. Their dielectric and piezoelectric properties are excellent and they are promising candidates



for high frequency dielectric studies. Furthermore, both the orthorhombic and tetragonal tungsten-bronze structures, as shown in Figs. 2 and 3, exist as a function of composition in these systems.

Recently, we examined the high frequency dielectric properties of the bronze composition crystal, $\text{Sr}_2\text{KNb}_5\text{O}_{15}$, between 30 to 40 GHz and found that the permittivity in the polar axis decreases substantially from its dc value, indicating that a major contributor to the polarizability relaxes at low frequencies. However, the high frequency dielectric properties of $\text{Sr}_2\text{KNb}_5\text{O}_{15}$ in the directions perpendicular to the polar axis do not show a comparable dispersion. To better characterize these orientational differences, it is planned to study related tungsten-bronzes that exhibit both tetragonal and orthorhombic ferroelectric systems such as $\text{Pb}_{1-2x}\text{K}_x\text{La}_x\text{Nb}_2\text{O}_6$ and $\text{Pb}_{1-x}\text{Ba}_x\text{Nb}_2\text{O}_6$. In the orthorhombic phase, there are generally two polar directions available, namely the b- and c- axes.

Initially, all measurements will be performed using ceramic disks and once the high frequency data is satisfactorily established, efforts will be expanded to grow bulk single crystals of suitable compositions exhibiting orthorhombic and tetragonal structures (tungsten bronze). Our current phase diagram work on these selected systems indicate that compositions from both of the systems melt congruently without losing appreciable amounts of lead. Once the crystals are available, we will study the directional and temperature dependence of the low and high frequency dielectric properties in both the orthorhombic and tetragonal crystal forms.

2.4 Perovskite $\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$ System

Single crystals of perovskite structure ferroelectric are difficult to grow and process in a single domain configuration. In all materials, both pure ferroelectric and partial ferroelectric:ferroelastic domains occur due to the very high symmetry ($m3m$) of the prototype paraelectric phase. Since Gibbs functions are available for BaTiO_3 , KNbO_3 , $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ and $\text{PbZr}_{1-x}\text{Ti}_x\text{O}_3$ systems, one can calculate the dielectric saturation functions for the single

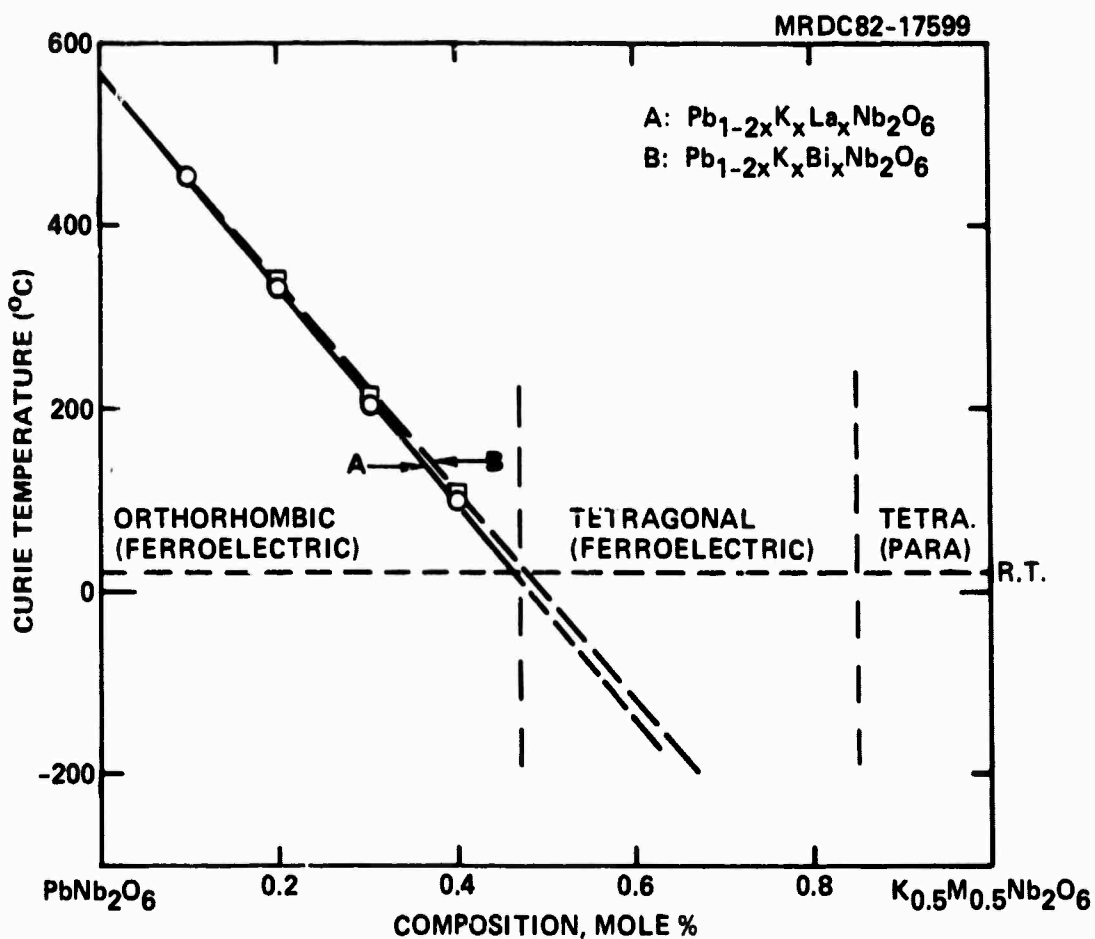


Fig. 2 Variation of ferroelectric transition temperature for $Pb_{1-2x}K_xM_xNb_2O_6$, M = La or Bi.

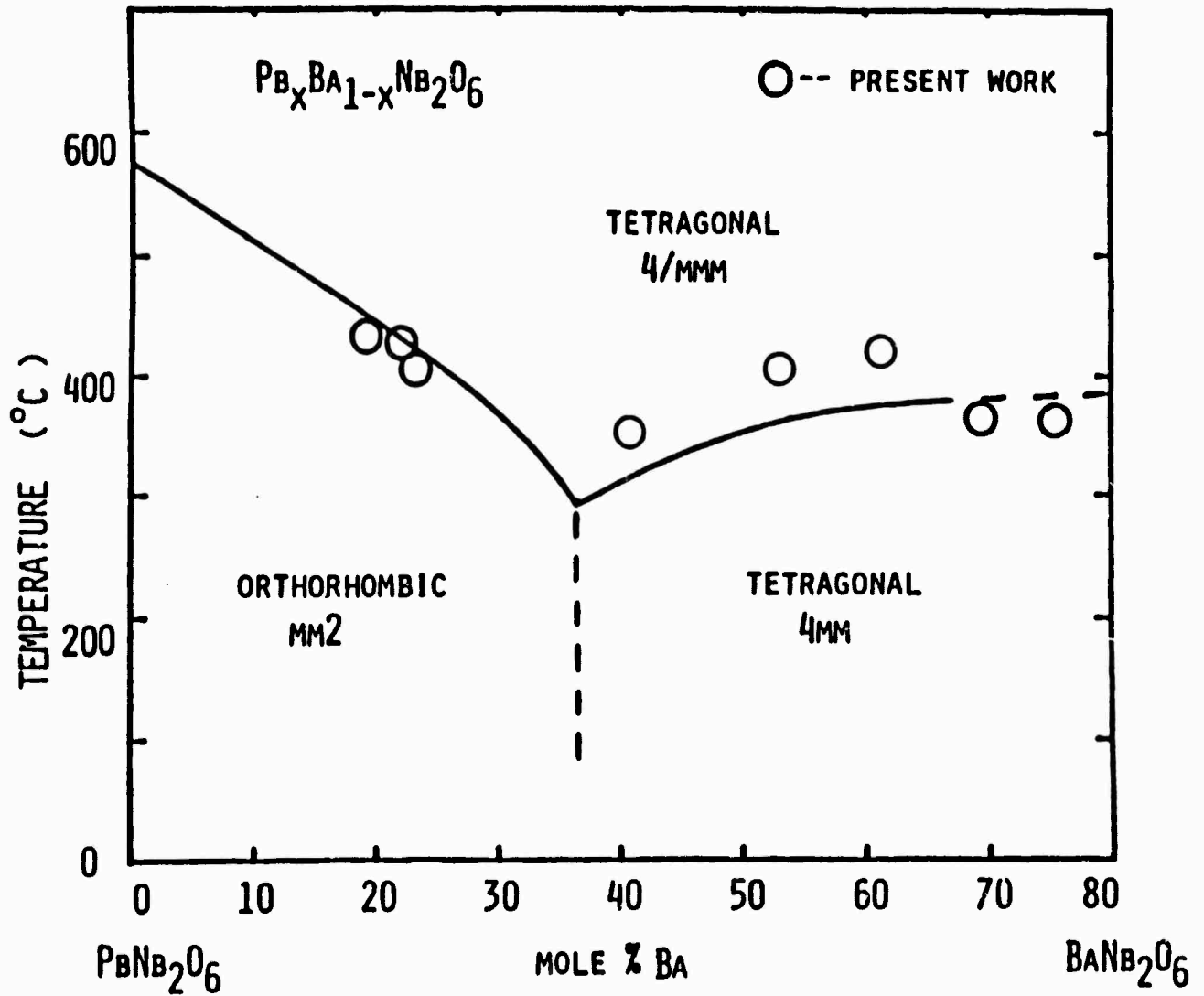


Fig. 3 Phase diagram for ferroelectricity in the solid solution system $Pb_{1-x}Ba_xNb_2O_6$.



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domain state in these families. We do not, however, anticipate a major advantage over the tungsten-bronze structural family.

The feature in the perovskite family which is of major interest is the multiaxial character of the ferroelectric response, which leads to interesting and useful dielectric, piezoelectric and electro-optic properties in the ceramic form. For the polar ferroelectric phases, the complex domain structure, grain-to-grain constraints, the highly anisotropic nature of the single domain permittivity, and the difficulty of processing to a perfect single phase assemblage may make interpretation of the response difficult. In the paraelectric phase, however, many of these difficulties are eliminated, and we expect that the quadratic response will be of more interest.

Ceramics based on the $Ba_{1-x}Sr_xTiO_3$ solid solution system exhibit both ferroelectric and paraelectric phases, and also exhibit very low microwave dielectric losses ($\tan \delta < 10^{-3}$). This fact makes these compositions attractive for the study of their high frequency dielectric properties in the present research work. The use of these compositions is well-known in the manufacture of ceramic capacitors and thermistors. Mixed titanates are also considered for other applications such as delay lines (SAW), slow wave structures and optical modulators.

In the present study, the composition of $Ba_{.7}Sr_{.3}TiO_3$ has been selected, since this composition has a room temperature ferroelectric phase transition and exhibits very low dielectric losses. Initially, high frequency dielectric properties will be studied on ceramic samples, and should the results appear favorable, efforts will be made to grow small single crystals in order to examine their directional and temperature dependence. The ceramic samples have been prepared by shaping and firing ceramic disks of the appropriate size from mixtures of $BaTiO_3$ and $SrTiO_3$ powders. All of the compositions were fired at $1450^\circ C$. Figures 4 and 5 show the dielectric constant and dielectric loss behavior for the $Ba_{1-x}Sr_xTiO_3$ composition. As can be seen from this data, the selected composition appears to be suitable and should be studied in detail. At present, work is underway to establish their high frequency dielectric properties at room temperature.

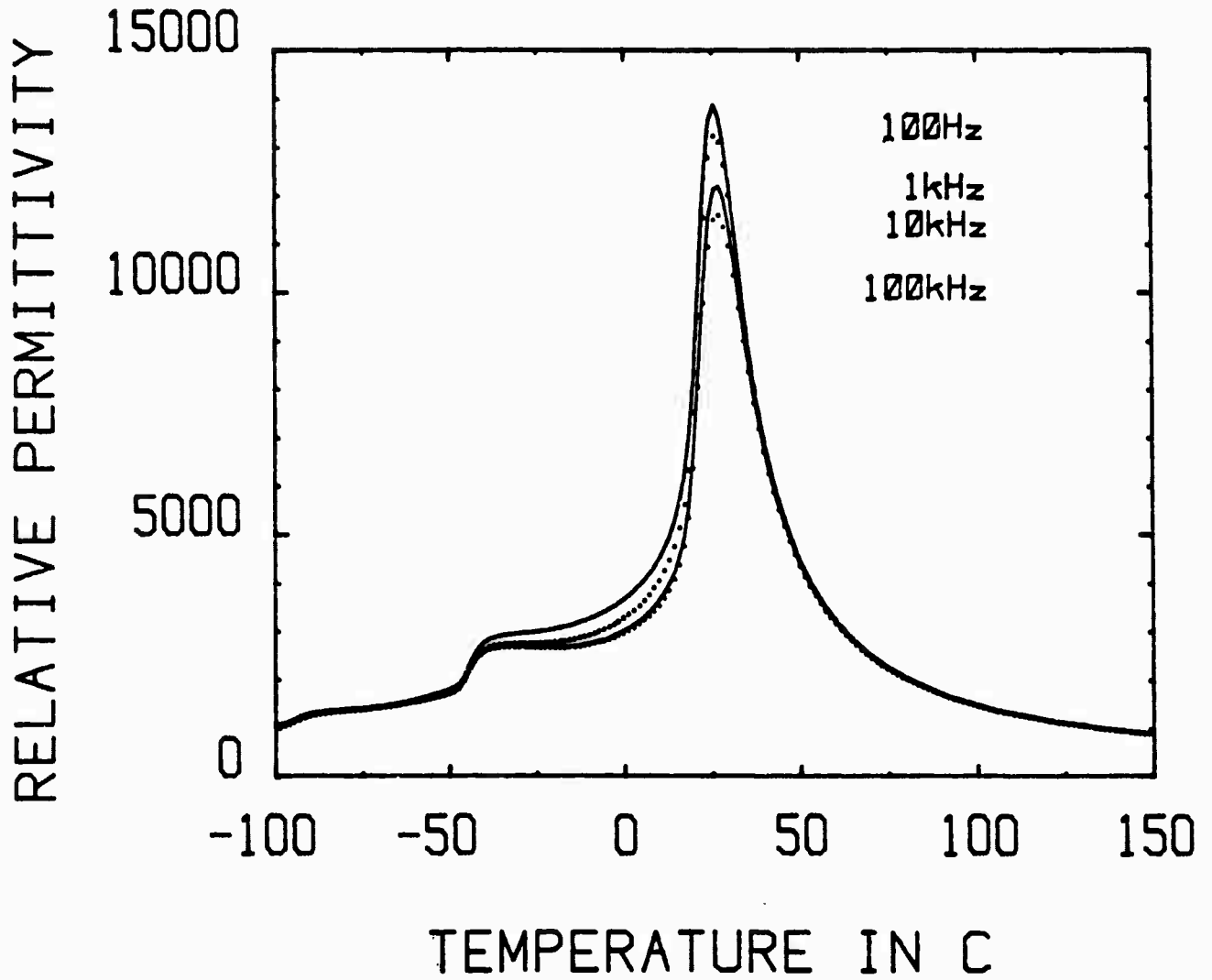


Fig. 4 Relative permittivity as a function of temperature for $Ba_{.70}Sr_{.30}TiO_3$.

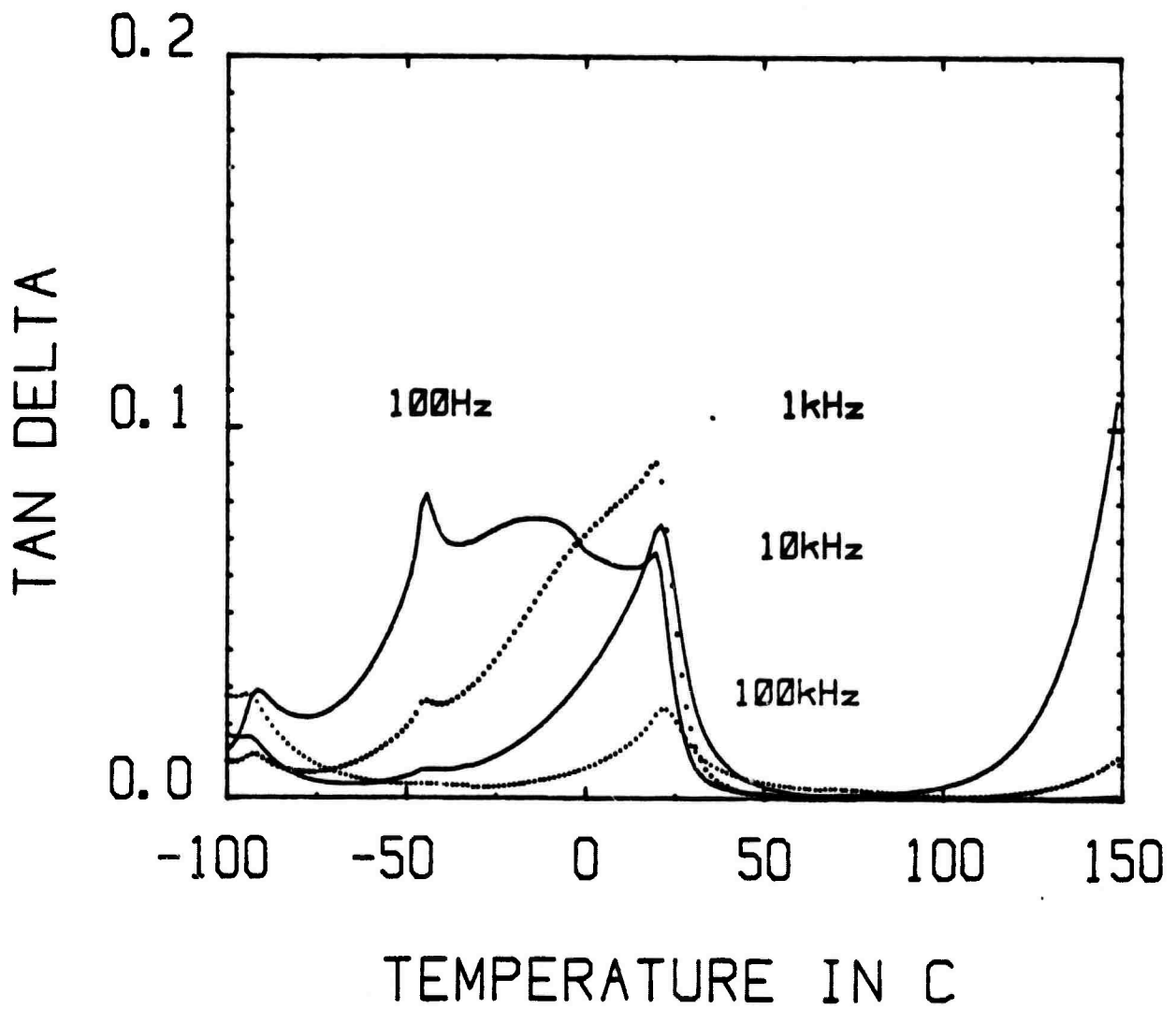


Fig. 5 $\text{Tan } \delta$ as a function of temperature for $\text{Ba}_{.70}\text{Sr}_{.30}\text{TiO}_3$.



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2.5 Future Materials

Several other important ferroelectric family crystals have been selected for future studies and they are as follows:

1. Single Crystal Bismuth Titanate - Bismuth titanate $\text{Bi}_4\text{Ti}_3\text{O}_{12}$ with $T_c = 675^\circ\text{C}$ shows unusual monoclinic dielectric properties in the ferroelectric single domain. Tilting of the optical indicatrix can be accomplished by domain switching - a similar effect should be possible at microwave frequencies.
2. Gadolinium Molybdate - $\text{Gd}_2(\text{MoO}_4)_3$ is the prototype for many improper ferroelectrics. Permittivity is low (~ 14) and should be largely dispersion-free, but again, K_{11} and K_{22} can be interchanged by domain switching.



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3.0 HIGH FREQUENCY DIELECTRIC MEASUREMENTS

3.1 SBN:60 Crystals

In order to explore the dependence of the observed high dielectric loss on growth conditions, a number of samples grown from ultra-pure chemicals were fabricated. Reflection and transmission measurements were carried out in waveguide for 30 to 40 GHz and from 90 to 100 GHz, and as a function of temperature on one polar axis sample.

In two samples cut to measure permittivity perpendicular to the polar axis, a dramatic decrease in loss was found near 40 GHz, with $\tan \delta_{11} < .03$, compared to a value of $\tan \delta_{11} = 0.1$ for the same samples at 30 GHz. One of these samples was subsequently cut down for measurement from 90 to 100 GHz, where $\tan \delta_{11}$ was found to have returned to values around 0.1.

A general feature of these measurements is a wide sample-to-sample variability, much as was noted in samples grown earlier in the program. Losses along the polar axis ranging from $\tan \delta_{33} = 0.2$ to 0.8 have been observed at a single frequency, indicating that extrinsic factors are dominating. Further effort on this system should, therefore, be directed toward the identification and control of these factors.

Measurements in the vicinity of the Curie point on a polar axis sample revealed a very broad peak in the loss with the position of the peak appearing to shift $\pm 10^\circ\text{C}$ over a relatively narrow range of frequency. Figure 6 shows the imaginary part of the relative permittivity $\epsilon'' = \epsilon' \tan \delta$ from 30°C to 140°C at three frequencies: 30, 33, and 36 GHz. The peak location is qualitatively consistent with the trend of low frequency measurements made at Penn State, which showed Curie temperatures in the ranges of 78°C to 85°C .

In this series of measurements, the behavior of the real part of the relative permittivity, as deduced from the reflection and transmission data, was surprisingly flat. Figure 7 shows the variation in ϵ' with temperatures at the same three frequencies selected in Fig. 6. The lack of any detectable peak in ϵ' near the Curie point most likely indicates a limitation on the

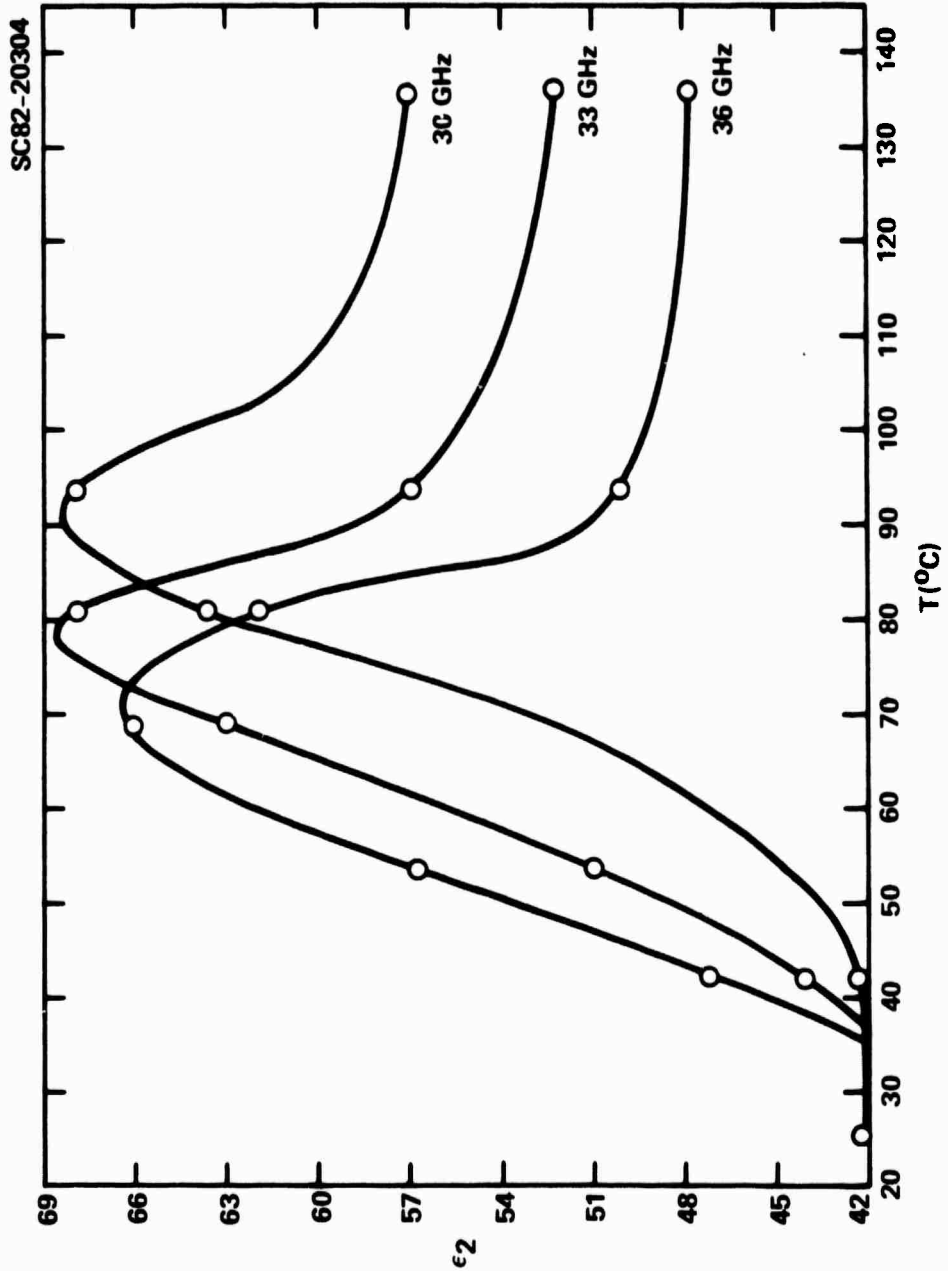


Fig. 6 Temperature dependence of imaginary relative permittivity at 30, 33, and 36 GHz.

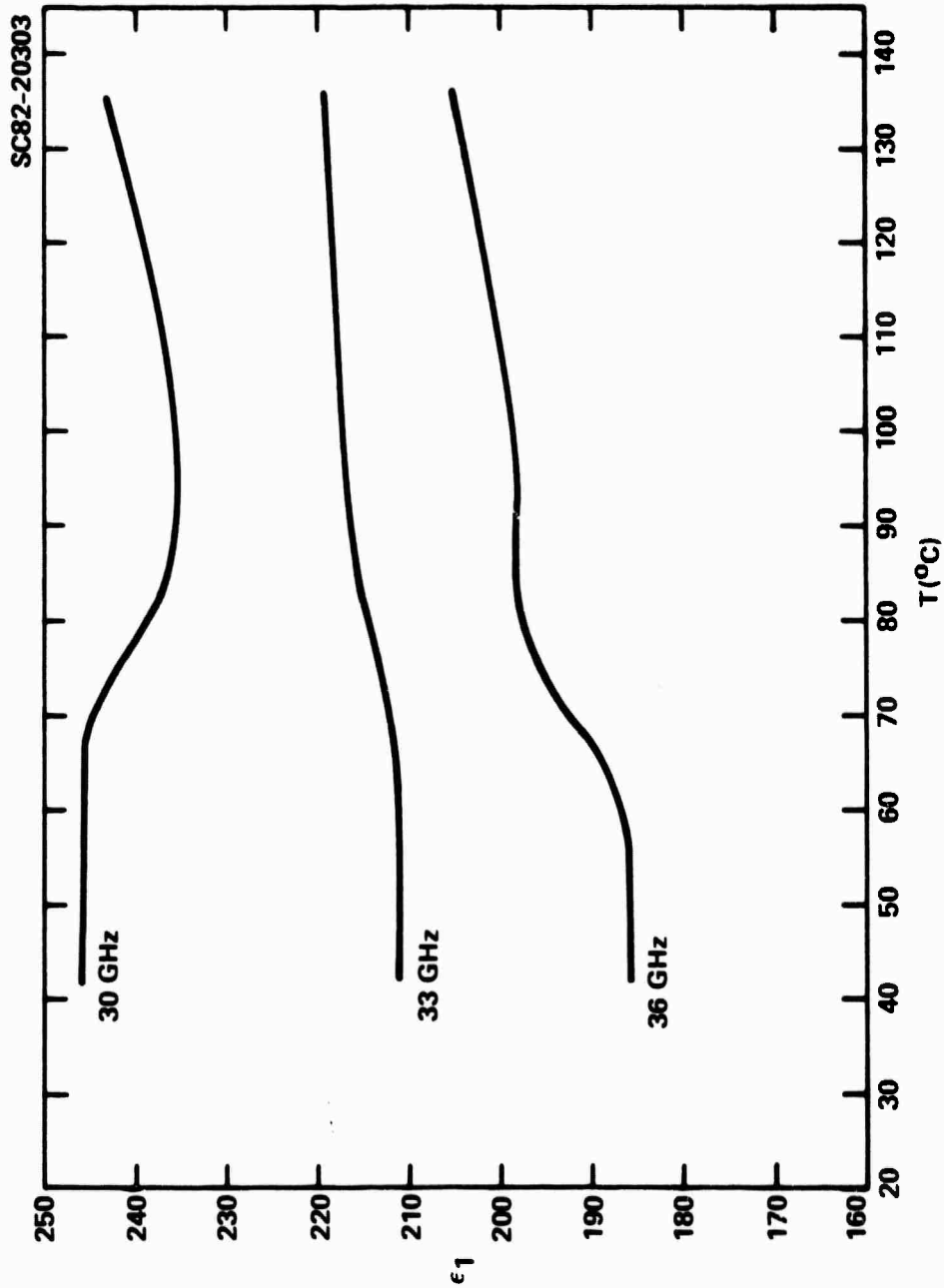


Fig. 7 Temperature dependence of real relative permittivity at 30, 33, and 36 GHz.



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measurement method, rather than a real property of the samples. A saturation phenomenon of this kind has been reported in the literature,¹⁷ and we are currently investigating whether similar limitations apply to our measurement configuration.

In view of this question, it is worthwhile to note that the values of ϵ'' deduced from these measurements have been found to be relatively insensitive to the measured value of ϵ' . This is because in high loss samples, ϵ'' is primarily determined by the transmission coefficient, while ϵ' is most sensitive to the reflection coefficient.

3.2 Lead Potassium Lanthanum Niobate (PKLN) System

Initial measurements have been carried out on orthorhombic PKLN ceramic samples from 30 to 40 GHz. PKLN exists in both orthorhombic and tetragonal forms, depending on the Pb:K ratio, and it is expected that the high frequency dielectric properties will show significant variation with composition and crystal symmetry. Depending upon the results of these investigations, single crystal growth of favorable compositions may be undertaken.

In the orthorhombic phases, the spontaneous polarization has components along two crystal axes, which should give rise to interesting behavior in the cross-axis sensitivity of dielectric properties to an applied field.

3.3 Barium Strontium Titanate (BST) System

The primary interest in BST ceramics for this program is that very low dielectric loss is typically found for this system at low frequency in the paraelectric phase. The particular composition currently under study ($\text{Ba}_{1-x}\text{Sr}_x\text{TiO}_3$) undergoes the transition to paraelectric phase around 25°C, so one expects relatively high losses at room temperature. Our initial measurements from 30 to 40 GHz bear out this expectation, with $\tan\delta$ of order 0.2 ($\epsilon' \sim 500$, $\epsilon'' \sim 80$). A systematic study at temperatures between 50°C and 100°C is planned.



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4.0 FUTURE PLANS

The search for low-loss ferroelectric systems will continue as a high priority in the remainder of the contract period. Variations in loss and permittivity within SBN crystals will be related to composition and growth conditions as a first step toward uncovering the factors controlling these variations.

Measurements of dn/dE will be carried out on the most interesting ferroelectrics at 35 and 94 GHz, including correlation with the observed variability in sample dielectric properties. A key question is to what extent the electric field sensitivity of the index scales with the microwave index in the SBN system.

Another important issue is the utility of variable index materials in millimeter wave radar systems. It is planned to interact with the user community to identify the current needs in terms of millimeter wave discrete and distributed active devices, and to establish the materials criteria and the design of devices which can potentially meet these needs.

Efforts will also be extended to improve the quality of SBN:60 and SBN:50 single crystals in order to reduce dielectric losses at high frequencies. Other ferroelectric materials such as $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ and $\text{Gd}_2(\text{MoO}_4)_3$ will be studied.



5.0 PUBLICATIONS AND PRESENTATIONS

5.1 Publications

1. R.R. Neurgaonkar, W.K. Cory, W.W. Ho, W.F. Hall and L.E. Cross, "Tungsten-Bronze Family Crystals for Acoustical and Dielectric Applications," *Ferroelectrics* 38, 857, 1981.
2. W.W. Ho, W.F. Hall, R.R. Neurgaonkar, R.E. DeWames and T.C. Lim, "Microwave Dielectric Properties of $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ Single Crystals at 35 and 58 GHz," *Ferroelectrics* 38, 833, 1981.
3. R.R. Neurgaonkar, J.R. Oliver, W.K. Cory and L.E. Cross, "Structural and Dielectric Properties for $\text{Pb}_{1-2x}\text{K}_x\text{M}_x^{3+}\text{Nb}_2\text{O}_6$ Phase, M = La or Bi," submitted to *Mat. Res. Bull.*

5.2 Presentations

1. R.R. Neurgaonkar, W.K. Cory, W.W. Ho, W.F. Hall and L.E. Cross, "Tungsten-Bronze Family Crystals for Acoustical and Dielectric Applications," presented at the 5th Int. Meeting on Ferroelectricity, Penn State Univ., PA, August 17-21, 1981.
2. W.W. Ho, W.F. Hall, R.R. Neurgaonkar, R.E. DeWames and T.C. Lim, "Microwave Dielectric Properties of $\text{Sr}_{0.61}\text{Ba}_{0.39}\text{Nb}_2\text{O}_6$ Single Crystals at 35 and 58 GHz," presented at the 5th Int. Meeting of Ferroelectricity at Penn State Univ., PA, August 17-21, 1981.



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