Dielectric Resonance in Ferroelectric Titanates in the Microwave Region

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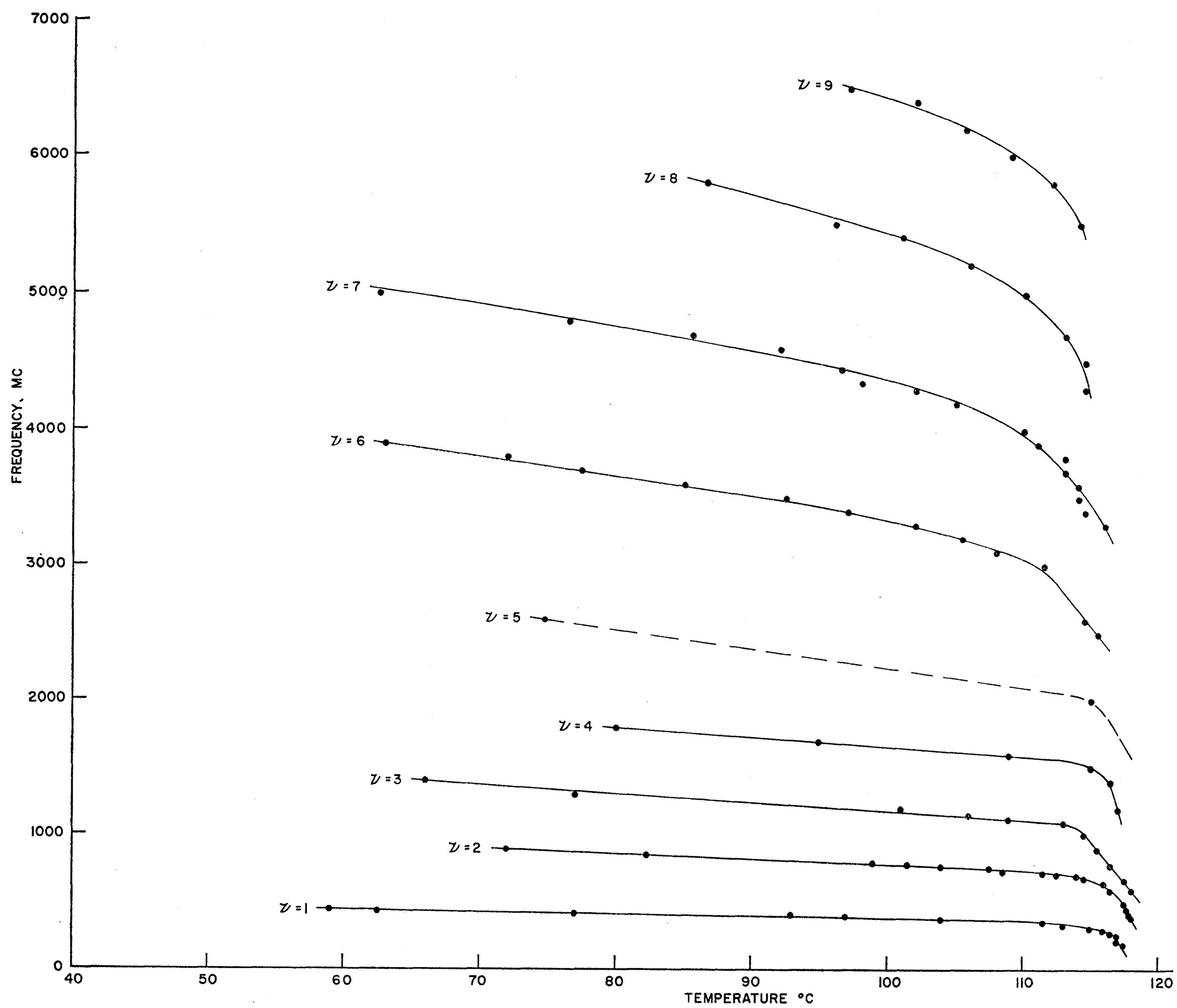
The dielectric spectra of $BaTiO_3$ and $(Ba\text{-}Sr)TiO_3$ have been measured in the microwave region from 250 Mc/sec to 7000 Mc/sec in the ferroelectric and paraelectric regions. The dispension near 2000 Mc/sec is shown to exhibit a resonance character near the Curie temperature.

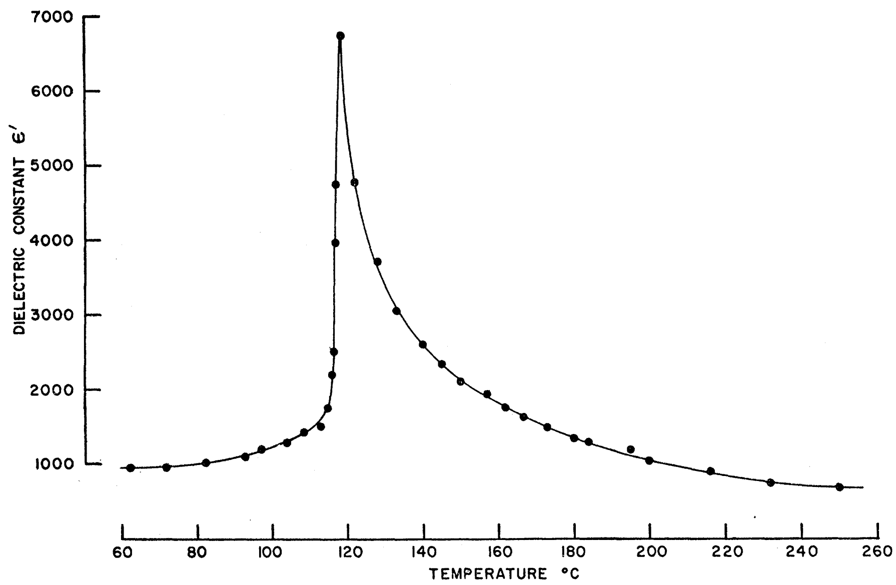
RECENT methods[1, 1a] have made it possible to observe the permittivity spectrum of ferroelectric materials as a function of temperature in an essentially continuous manner and as a function of frequency over a large portion of the microwave region. The work of Powles and Jackson,[2] von Hippel and Westphal,[3] and Fousek[4] has indicated that a relaxation occurs in the permittivity of barium titanate near 2000 Mc/sec. Here is reported the results of measurements of the permittivity of $BaTiO_3$ and $(Ba\text{-}Sr)TiO_3$ from 250 Mc/sec to 7000 Mc/sec over a temperature range from 30° to 260°C.

It will be seen that not only a relaxation is observed, but a more general dispersion is apparent near the Curie



(a)

[1] G. Rupprecht, Scientific Report No. 1, AFCRC-TN-596, Research Division, Raytheon Company, 1960 (unpublished).
[1a] *Note added in proof.* G. Rupprecht, R. O. Bell, and B. D. Silverman, Phys. Rev. **123**, 97 (1961).
[2] J. G. Powles and W. Jackson, Proc. Inst. Elec. Engrs. (London), **96**, Pt III, 383 (1949).
[3] A. von Hippel and W. G. Westphal, National Research Council Conference on Electrical Insulation, October, 1948 (unpublished).
[4] J. Fousek, Czech. J. Phys. **9**, 172 (1959).



(b)

FIG. 1. (a) Transmission frequencies of a polycrystalline $BaTiO_3$ sample plotted as a function of temperature in the ferroelectric region. (b) Dielectric constant of $BaTiO_3$ as a function of temperature at about 1000 Mc/sec. This curve is a composite of data calculated from curves similar to Fig. 1(a).

temperature. The method of measurement, due to Rupprecht,[1] consists of observing the transmission of microwave energy through a ferroelectric sample in a coaxial line. In general, most of the microwave energy is reflected at the sample face and little or no transmission occurs. However, when the optical path length of the sample $(\epsilon')^{\frac{1}{2}}d$, where d is the sample length and ϵ' is the real part of the dielectric constant, is an integral number of free-space half-wavelengths, or

$$(\epsilon')^{\frac{1}{2}}d=\nu\lambda_0/2, \qquad (1)$$

the sample acts like a transmission cavity and a sizeable transmitted signal is observed. The sample is heated and allowed to cool slowly. A thermocouple near the sample provides the X input to a recorder and the detected transmitted signal is the Y input. The process is repeated at different frequencies, and a plot of the frequency vs temperature at which transmission occurs is then a field of points, any one of which may be used to calculate the dielectric constant according to (1).

Figure 1(a) shows the field of points obtained in the ferroelectric region for polycrystalline $BaTiO_3$. Figure 1(b) shows the dielectric constant as a function of temperature that results from these data and similar

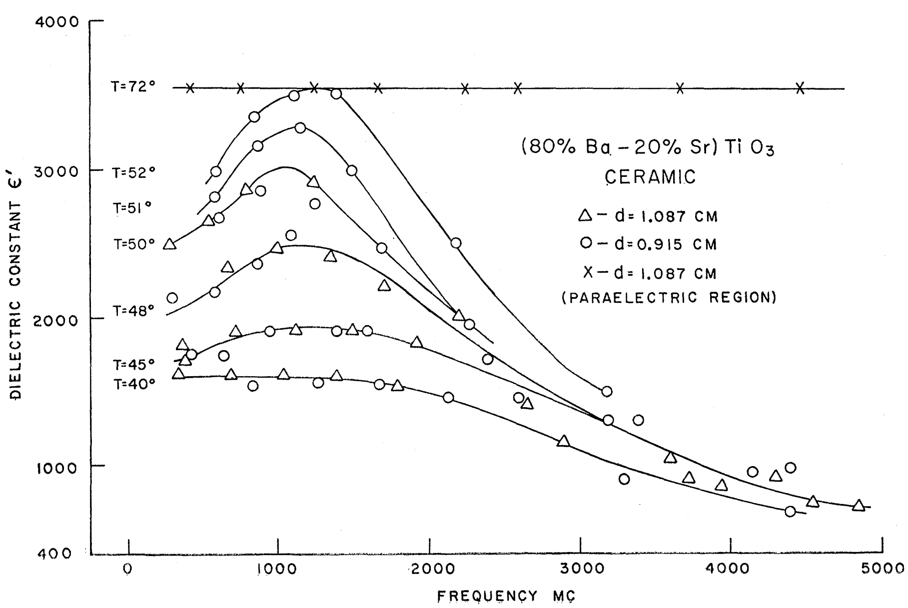


FIG. 2. The dielectric spectrum of (80%Ba–20%Sr)TiO_3 at temperatures near the Curie point (55°C).

data obtained in the region above the Curie point (122°C). Similar data were obtained for (80%Ba-20%Sr)TiO_3.

The dispersion is best displayed by showing the permittivity as a function of frequency. Figure 2 shows the dispersion observed using polycrystalline (Ba-Sr) TiO_3 samples (Curie temperature 55°C). Note that the dispersion is not a simple relaxation process, but exhibits resonance characteristics as the Curie point is approached from the ferroelectric side. No dispersion is apparent above the Curie point; one curve in this region is also shown in Fig. 2. The data displayed in Fig. 2 were obtained using two samples of incommensurate lengths of the same material. The appearance of resonance character in the permittivity spectrum recalls Kittel's theory of domain boundary inertia.[5] It appears that the domain walls respond in a manner similar to a forced damped harmonic oscillator.

Further measurements are in progress to determine the effect of biasing electric fields on the dispersion of (Ba-Sr)TiO_3 ferroelectrics. The effect of grain size on the spectrum is also of interest to examine the possibility of acoustical resonance.

[5] C. Kittel, Phys. Rev. **83**, 458(1951).

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Antiferromagnetic Susceptibility of the Plane Triangular Ising Lattice

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The magnetic moment transformation developed by Fisher enables the antiferromagnetic susceptibility of the plane triangular Ising lattice to be expanded as a power series that converges over the whole temperature range $0\leqslant T\leqslant\infty$. The dominant asymptotic behavior of the coefficients conjectured from extrapolations by Domb and Sykes, and independently by Park, has been established theoretically by Fisher. A counting theorem based on the method of Oguchi enables the first twelve terms of the expansion to be derived. It is found possible to evaluate the susceptibility numerically over the whole temperature range with a maximum error of 0.1% at $T=0$. It is concluded that the specific susceptibility per spin ($kT\chi_0/m^2$) falls smoothly from unity at $T=\infty$ to a value at $T=0$ which does not differ by more than 0.1% from 5/36, and the form of the counting theorem leads it to be surmised that it is exactly 5/36.

1. INTRODUCTION

NO exact closed analytical expression has yet been given for the zero-field susceptibility of any two-dimensional Ising model. It is the purpose of the present paper to evaluate the antiferromagnetic susceptibility of the plane triangular lattice numerically from exact series developments.

At high temperatures the reduced zero-field susceptibility χ defined as $kT\chi_0/m^2$ may be expanded in powers of the high-temperature counting variable $v=\tanh(J/kT)$, by the method of Oguchi[1,2] as

$$\chi(v)=\sum_n a_n v^n,\quad a_0=1. \tag{1}$$

The first 12 coefficients have been obtained by Sykes.[2] The a_n are positive integers and the series converges[3] only for $|v|\leqslant v_f$, where $v_f=\tanh(J/kT_f)=0.267949$ and T_f is the ferromagnetic Curie temperature. The series (1) can only be used to estimate $\chi(v)$ in the range $|v|\leqslant v_f$, i.e., $T_f\leqslant T\leqslant\infty$.

[1] T. Oguchi, J. Phys. Soc. Japan **6**, 31 (1951).
[2] M. F. Sykes, J. Math. Phys. **2**, 52 (1961).
[3] C. Domb and M. F. Sykes, J. Math. Phys. **2**, 63 (1961).

At $T=0$ which corresponds to $v=-1$ the ground state of the antiferromagnetic triangular lattice is highly degenerate[4] and no series or asymptotic developments about this origin have so far been given.

Recently Fisher[5] has developed a magnetic moment transformation that relates the reduced susceptibility of the triangular lattice to that of the honeycomb lattice. If χ_T denotes the reduced susceptibility of the triangular lattice and χ_H that of the honeycomb lattice, then

$$\chi_T(v)=\tfrac{1}{2}[\chi_H(w)+\chi_H(-w)], \tag{2}$$

for

$$w^2=v(1+v)/(1+v^3). \tag{3}$$

Equation (2) relates the susceptibility of the triangular lattice at temperature v to the mean of the ferromagnetic and antiferromagnetic susceptibilities of the honeycomb lattice at a temperature w determined by (3). As v varies from 0 to -1, w^2 varies from 0 to $-\frac{1}{3}$ and the high-temperature honeycomb susceptibility series corre-

[4] G. H. Wannier, Phys. Rev. **79**, 357 (1950).
[5] M. E. Fisher, Phys. Rev. **113**, 969 (1959).