

The agreement between the predicted and measured results is excellent. For Al-Ag where  $Y_{Al}/Y_{Ag} \approx 1$ , Eqs. (6) and (7) are formally equivalent; the fitting of the data would not be significantly altered if the Ag layers were the main sources of new dislocations.

In conclusion, it has been established that the observed strength enhancement in thin-layered Al-Cu and Al-Ag laminates is caused by the repulsive dislocation-image forces described by Koehler.<sup>1</sup> The laminate systems investigated consisted of polycrystalline layers with nearly random grain orientations, and consequently the individual layers present rather complex three-dimensional slip systems. The calculated results are therefore approximate for they do not include the rigorous angular dependences of the forces acting in the possible glide planes.

The author is indebted to R. L. Wyatt for his invaluable technical assistance in the experimental

work and is also greatly thankful to C. R. Whitsett and D. P. Ames of the McDonnell Douglas Research Laboratories and W. T. Highberger of the Naval Air Systems Command for their many illuminating discussions of the problem. This research was supported by the U. S. Department of the Navy, Naval Air Systems Command.

<sup>1</sup>J. S. Koehler, Phys. Rev. B 2, 547 (1970).

<sup>2</sup>A. K. Head, Proc. Phys. Soc., London, Sect. B 66, 793 (1953), and Philos. Mag. 44, 92 (1953).

<sup>3</sup>R. E. Schmunk and C. S. Smith, J. Phys. Chem. Solids 9, 100 (1959).

<sup>4</sup>W. C. Overton, Jr., and J. Gaffney, Phys. Rev. 98, 969 (1955).

<sup>5</sup>R. Bacon and C. S. Smith, Acta Metall. 4, 337 (1956).

<sup>6</sup>W. J. Petch, J. Iron Steel Inst. 173, 25 (1953).

<sup>7</sup>J. Friedel, *Dislocations* (Addison-Wesley, Reading, Mass., 1964), p. 261.

<sup>8</sup>N. F. Mott, Philos. Mag. 43, 1151 (1952).

## Dynamic Scaling near the Lambda Point in Liquid Helium

Richard A. Ferrell

*Institute for Physical Science and Technology and Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742*

and

Volker Dohm

*Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742,<sup>(a)</sup> and Institut für Festkörperforschung, Kernforschungsanlage Jülich, 517 Jülich, Germany<sup>(b)</sup>*

and

Jayanta K. Bhattacharjee

*Department of Physics and Astronomy, University of Maryland, College Park, Maryland 20742*

(Received 5 July 1978)

Two-loop calculations indicate that the order parameter in liquid helium at the  $\lambda$  point may relax by an order of magnitude more slowly than the entropy. Consequently the critical region for entropy relaxation is expanded by an order of magnitude, both above and below the  $\lambda$  point, which may help to reconcile light-scattering data, second-sound damping, and dynamic scaling theory. Characteristic double-humped spectra are calculated, providing a crucial test of the present theory.

From general considerations a dynamic scaling theory<sup>1,2</sup> has been advanced in which it has been argued that the breadth of the spectrum of entropy fluctuations at the  $\lambda$  point in liquid helium scales with wave number  $k$  according to

$$\omega_c(k) = ak^{3/2}. \quad (1)$$

Equation (1) has subsequently been confirmed by both the mode-coupling<sup>3</sup> and dynamic renormalization-group theories.<sup>4</sup> The value of the con-

stant  $a$  which is obtained from these theories compares satisfactorily with the value determined experimentally from light scattering by Winterling, Holmes, and Greytak,<sup>5</sup> Vinen *et al.*,<sup>6</sup> and Tarvin, Vidal, and Greytak.<sup>7</sup> The temperature dependence predicted by dynamic scaling has, however, been found to be in gross contradiction with the experimental observations. In this note we offer an explanation of this discrepancy and show how dynamic scaling theory can

be modified so as to bring it into agreement with experiment for liquid helium.

The entropy current is a consequence of the fluctuation in the complex order parameter  $\psi = \psi_1 + i\psi_2$  and is given by an expression of the usual quantum mechanical form

$$\vec{J} \propto -\frac{1}{2}i(\psi^* \nabla \psi - \psi \nabla \psi^*). \quad (2)$$

From the continuity equation, the rate of change of the entropy density  $S$  is proportional to the divergence of Eq. (2). Consequently the rate of change of the Fourier component associated with wave number, or "momentum,"  $k$ , is

$$\dot{S}_k \propto \sum_{p, p'} (p^2 - p'^2) \psi_1(\vec{p}) \psi_2(\vec{p}') \quad (3)$$

with the sum restricted by momentum conservation

$$\vec{k} = \vec{p} + \vec{p}'. \quad (4)$$

Thus it is important to bear in mind that the dynamics of an entropy fluctuation at momentum  $k$  are not directly determined by  $k$ , but instead by the range of values of the momenta  $p$  and  $p'$ . Normally the values of  $p$  and  $p'$  that contribute most heavily to a convolution such as that of Eq. (3) would be expected to be of the order of  $k$ . Our basic point is that this is probably not true in the present case. We pursue here the consequences that ensue from the possibility that the important momenta in Eq. (3) are in the range  $p \approx p' \gg k$ . This situation can arise when the frequency width of the order-parameter fluctuation spectrum  $\omega_\psi(k) \ll \omega_c(k)$ . It is convenient to write

$$\omega_\psi(k) = \sigma_\psi \omega_c(k), \quad (5)$$

where the dimensionless ratio  $\sigma_\psi$  is a small number. Because of the unobservability of  $\omega_\psi(k)$  there is no direct experimental indication of the smallness of  $\sigma_\psi$ . An indication from theory, however, is the observation of De Dominicis and Peliti<sup>8</sup> in two-loop order that an instability may occur in which  $\sigma_\psi$  vanishes. The stability question has been studied in detail<sup>9,10</sup> by Dohm and Ferrell and in a slightly different way by Ferrell and Bhattacharjee.<sup>11</sup> In both of these treatments small, positive values of  $O(10^{-1})$  are found for  $\sigma_\psi$ . Therefore it seems appropriate and worthwhile to explore here the consequences ensuing from the assumption

$$\sigma_\psi \ll 1. \quad (6)$$

The improved agreement with experiment that we will achieve serves to indicate the correctness of Eq. (6).

For the sake of simplicity we first describe qualitatively the consequences of Eq. (6). As we shall discuss more explicitly below, an order-parameter fluctuation of momentum  $p$  only contributes effectively to the momentum- $k$  entropy fluctuation when its frequency matches that of the latter. Therefore we determine the important range of  $p$  by requiring

$$\omega_\psi(p) = \omega_c(k). \quad (7)$$

Substitution from Eqs. (6) and (1) then yields

$$p = \sigma_\psi^{-2/3} k. \quad (8)$$

Applying now the basic dynamic scaling idea to  $p$  instead of to  $k$ , we see that we should not expect to observe a temperature effect in the spectrum of  $S_k$  until the inverse correlation length attains the value  $\kappa \approx p$ . According to Eq. (8) this can be expressed as  $\bar{\kappa} \approx k$ , where

$$\bar{\kappa} = \sigma_\psi^{2/3} \kappa. \quad (9)$$

This is our main result and shows that effectively the correlation length controlling the dynamics is increased by the factor  $\sigma_\psi^{-2/3}$ . Therefore *the temperature width of the dynamic critical region is expanded by  $\sigma_\psi^{-1}$  or roughly one order of magnitude, according to the theoretical  $\epsilon$ -expansion estimates cited above.*

In order to demonstrate in detail how the above ideas work out, we examine now some of the properties of the entropy self-energy,  $\gamma_s(k, \omega)$ , and in particular, its dependence on frequency  $\omega$ . We first limit ourselves to the  $\lambda$  point where  $\bar{\kappa} = 0$ . It suffices to recall that in every order of perturbation theory (i.e., arbitrary number of loops) dimensional arguments yield for  $\gamma_s(k, 0)$  the  $k$  dependence  $k^2$  times  $k^{-1/2}$ . The latter factor comes essentially from cutting off the  $p$  integrals at  $p \approx k$ . On the other hand, at finite frequency, an additional term  $-i\omega$  occurs in the denominators of the integrals for  $\gamma_s(k, \omega)$ . For sufficiently large  $\omega$  the integrals are therefore frequency cut off and instead of being proportional to  $k^{-1/2}$  they are proportional to  $(-i\omega)^{-1/3}$ . These two limiting cases are expressed, respectively, by

$$\gamma_s(k, \omega) \propto k^2 \begin{cases} k^{-1/2} & (10a) \\ (-i\omega)^{-1/3} & (10b) \end{cases}$$

Let us consider the limiting case  $\sigma_\psi = 0$ . Then the entire spectrum falls in the "high-frequency" range, and Eq. (10b) applies throughout. By introducing the dimensionless frequency

$$\Omega = \omega / \omega_c(k) \quad (11)$$

we can write Eq. (10b) in the form

$$\gamma_S = \omega_c (-i\Omega)^{-1/3}. \quad (12)$$

This relates the coefficient  $a$  of Eq. (1) to the proportionality constant in Eq. (10b). Substitution of Eq. (12) gives us now the entropy Green's function

$$g_S(k, \omega) = 1/[-i\omega + \gamma_S(k, \omega)] \\ = \omega_c^{-1}/[-i\Omega + (-i\Omega)^{-1/3}]. \quad (13)$$

The spectral function, which is to be compared with the intensity of the light scattered at frequency shift  $\omega$ , is

$$\omega_c \text{Reg}_S(k, \omega) = \frac{\sqrt{3}}{2} \frac{\Omega^{1/3}}{\Omega^{3/3} - \Omega^{4/3} + 1}. \quad (14)$$

The sum rule

$$\int_{-\infty}^{\infty} \text{Re } g_S(k, \omega) d\omega = \pi \quad (15)$$

can be confirmed explicitly by substitution from Eq. (14). The spectrum of Eq. (14) is strongly non-Lorentzian. Its vanishing at  $\omega=0$  is a conse-

quence of the limit  $\sigma_\psi = 0$ . The central valley begins to fill in and becomes  $O(\sigma_\psi^{1/3})$  as  $\sigma_\psi$  takes on a small positive value. But for  $\sigma_\psi \ll 1$  this is a minor effect which leaves the wings of the spectrum essentially unchanged. Therefore we pass on to the more interesting question of the changes produced in the spectrum by finite values of  $\kappa$ .

We now carry out the single-loop convolution integral corresponding to the "decoupled-mode" version<sup>12</sup> of mode-coupling.<sup>3</sup> We have also determined the shape function to  $O(\epsilon)$ ,<sup>13</sup> but here we report on our results for the three-dimensional integral. We assume that the dependence of the order-parameter relaxation rate on  $p$  and  $\kappa$  is given by  $\sigma_\psi a(p^2 + \kappa^2)^{3/4}$ . As will be shown elsewhere, the somewhat more complicated form determined self-consistently in Ref. 14 leads to substantially the same results. For  $\kappa \gg k$  the dominant contribution comes from the momentum range  $p \gg k$ . This results in a high-momentum simplification of the convolution integral. All angular dependences then disappear, leaving only the radial integration

$$\gamma_S(k, \bar{\kappa}, \omega) = \omega_c k^{1/2} \frac{3^{3/2} a \sigma_\psi^{2/3}}{2^{4/3} \pi} \int_0^\infty dp \frac{[p^2/(p^2 + \kappa^2)]^2}{-i\omega + 2\sigma_\psi a(p^2 + \kappa^2)^{3/4}} = \omega_c \left(\frac{k}{\bar{\kappa}}\right)^{1/2} \frac{3^{3/2}}{2^{7/3}} I\left(\frac{-i\Omega}{2} \left(\frac{k}{\bar{\kappa}}\right)^{3/2}\right), \quad (16)$$

where

$$I(Z) = \int_0^\infty d\pi \frac{1}{Z + (\pi^2 + 1)^{3/4}} \left(\frac{\pi^2}{\pi^2 + 1}\right)^2. \quad (17)$$

Here we have introduced the dimensionless momentum  $\pi = p/\kappa$ . Equation (16) has been normalized so as to reduce to Eq. (12) in the limit  $\bar{\kappa}/k \rightarrow 0$ . In other words, because  $\omega_c$  is essentially the median frequency of the entropy fluctuation spectrum, the prefactors in Eq. (16) are required for consistency with Eq. (12). We further note at this point that the usual frequency ratio  $w$  of the order parameter and entropy relaxation rates is in the present notation of  $O(\sigma_\psi^{4/3})$ .

For  $|Z| \gg 1$  the asymptotic limit of Eq. (17) is

$$I(Z) \sim \int_0^\infty (Z + \pi^{3/2})^{-1} d\pi = (4\pi/3\sqrt{3}) Z^{-1/3}. \quad (18)$$

On the other hand, in the range  $|Z| \ll 1$ ,

$$I(Z) \simeq I(0)[1 + ZI'(0)/I(0)] \\ = I(0)[1 - \frac{1}{3}(-i\Omega/\sigma)], \quad (19)$$

where

$$\sigma = -\frac{2}{3} \frac{I(0)}{I'(0)} \left(\frac{\bar{\kappa}}{k}\right)^{3/2} = \frac{20}{21} \sqrt{\pi} \frac{\Gamma(\frac{1}{4})/\sqrt{\bar{\kappa}}}{\Gamma(\frac{3}{4})/\sqrt{k}} \\ = 5.0(\bar{\kappa}/k)^{3/2}. \quad (20)$$

Equations (18) and (19) suggest a convenient approximation to  $I(Z)$ , which when substituted into Eq. (17) becomes

$$\gamma_S(k, \bar{\kappa}, \omega) = \omega_c (\sigma - i\Omega)^{-1/3}. \quad (21)$$

Although Eq. (21) overestimates  $\gamma_S$  in the small-frequency range by approximately 20%, it gives a representation of the overall frequency dependence that is sufficiently accurate for the present purposes. The resulting curves for  $\omega_c \text{Reg}_S$  are plotted in Fig. 1 for the four different values of  $\sigma^{1/3}$  equal to  $\frac{1}{8}$ ,  $\frac{1}{2}$ , 1, and  $\sqrt{2}$ , as identified by the central value at  $\Omega=0$ . The shape is seen to vary from a two-Lorentzian flat form to that of a single Lorentzian, comparing satisfactorily with the experimental trend. With  $\sigma$  increasing, as a consequence of the growth of  $\bar{\kappa}/k$ , the center of the spectrum fills in, while the wings change very little. Only after the central valley is filled up, resulting in a nearly Lorentzian shape, does the spectrum start to shrink in the manner expected from conventional dynamic scaling.<sup>14</sup> A convenient measure of the frequency width of the spectrum is the median frequency,<sup>2</sup> which is plotted as the lower curve in Fig. 2. The expansion of the spectral critical region predicted by Eq. (9)

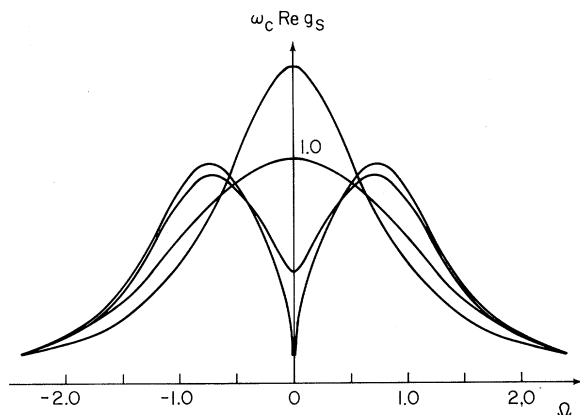


FIG. 1. Spectral strength of entropy fluctuations vs frequency  $\Omega$  for  $\sigma^{1/3} = \frac{1}{8}, \frac{1}{2}, 1$  and  $\sqrt{2}$ , as identified by the strength at  $\Omega = 0$ . Raising the temperature above the  $\lambda$  point increases  $\sigma$  in a linear proportion.  $\sigma < 1$  and  $\sigma > 1$  correspond to the (median frequency) critical and hydrodynamic regions, respectively. The spectral strength is obtained from the real part of Eq. (13) after substituting Eq. (21).

is clearly evident. In contrast is the upper curve of Fig. 2 which shows the zero-frequency value of the entropy relaxation rate,  $\gamma_S/\omega_c$ . This is the quantity that determines the temperature dependence of the thermal conductivity, and its critical region is unaffected by the present considerations.

The above considerations above the  $\lambda$  point can also be applied below the  $\lambda$  point. Although the dynamics of the ordered state are more complicated, we expect that the damping of second sound in the critical region is determined by the high-frequency tail of  $\gamma_S$ . Evaluated at the propagating frequency  $\omega_2 \propto k\kappa^{1/2}$ , Eq. (10b) predicts a breadth proportional to  $\kappa^{-1/6}$ . The corresponding exponent of  $-\frac{1}{9}$  for the temperature dependence may be sufficiently small to be consistent with the reported constancy of the width of the second sound doublet lines. We expect the  $-\frac{1}{9}$  power law to hold over a critical-temperature region expanded by the factor  $\sigma_\psi^{-3/2}$ . Finally, with further lowering of the temperature, the conventional  $-\frac{1}{3}$  power sets in in the hydrodynamic region. Because of the expansion of the critical region, the strength of the damping in the hydrodynamic region is enhanced by  $\sigma_\psi^{-1/3}$ , in the scale of  $\omega_c$ . This may account, at least partly, for the factor of five<sup>14,15</sup> by which Tyson's measured second-sound damping<sup>16</sup> exceeds the estimate based on conventional theory. These speculations on the situation below the  $\lambda$  point are obviously not com-

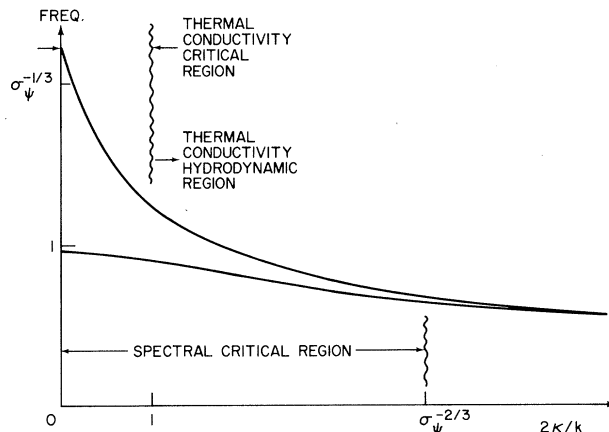


FIG. 2. Two different characteristic frequencies for the entropy fluctuation spectrum (versus inverse correlation length in units of  $\frac{1}{2}k = \pi/\lambda$ , where  $\lambda$  is the fluctuation wavelength). The upper curve shows the zero-frequency value of the entropy relaxation rate,  $\gamma_S/\omega_c$ , which has the conventional critical region as shown. The lower curve exhibits the median frequency of Halperin and Hohenberg (Ref. 2) for spectra of the type shown in Fig. 1. The critical region for the median frequency is expanded by the factor  $\sigma_\psi^{-2/3}$ . The curves have been calculated for  $\sigma_\psi = \frac{1}{8}$ .

elling and will require close quantitative study.

To summarize, we have presented a simple physical argument which accounts for the enlarged critical region found by Tarvin, Vidal, and Greytak.<sup>7</sup> The explanation relies upon the presumed very slow relaxation of the order parameter.<sup>17</sup> In addition, we have predicted in Fig. 1 the changes expected in the spectral shape as liquid helium is brought out of the critical region. The double-hump shape ought to persist over an appreciable temperature interval above the  $\lambda$  point. This would provide a crucial test of our theory if the experimental resolution could be increased sufficiently.

This work has been supported in part by the U. S. National Science Foundation.

<sup>(a)</sup> Present address.

<sup>(b)</sup> Permanent address.

<sup>1</sup>R. A. Ferrell, N. Menyh ard, H. Schmidt, F. Schwabl, and P. Sz epfalusy, Phys. Rev. Lett. **18**, 891 (1967), and Ann. Phys. (N.Y.) **47**, 565 (1968).

<sup>2</sup>B. I. Halperin and P. C. Hohenberg, Phys. Rev. **177**, 952 (1969).

<sup>3</sup>K. Kawasaki, Ann. Phys. (N.Y.) **61**, 1 (1970).

<sup>4</sup>B. I. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. B **13**, 1299 (1976); P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).

<sup>5</sup>G. Winterling, F. S. Holmes, and T. J. Greytak, *Phys. Rev. Lett.* **30**, 427 (1973).

<sup>6</sup>W. F. Vinen, C. J. Palin, J. M. Lumley, D. L. Hurd, and J. M. Vaughn, in *Low Temperature Physics, LT-14*, edited by M. Krusius and M. Vuorio (North-Holland, Amsterdam, 1975), Vol. I, p. 191.

<sup>7</sup>J. A. Tarvin, F. Vidal, and T. J. Greytak, *Phys. Rev. B* **15**, 4193 (1977).

<sup>8</sup>C. De Dominicis and L. Peliti, *Phys. Rev. Lett.* **38**, 505 (1977), and *Phys. Rev. B* **18**, 353 (1978).

<sup>9</sup>V. Dohm and R. A. Ferrell, University of Maryland Technical Report No. 78-077 (unpublished), and, *Phys. Lett.* **67A**, 387 (1978).

<sup>10</sup>V. Dohm, to be published.

<sup>11</sup>R. A. Ferrell and J. K. Bhattacharjee, University of Maryland Technical Report No. 78-080 (unpublished), and to be published.

<sup>12</sup>R. A. Ferrell, *Phys. Rev. Lett.* **24**, 1169 (1970).

<sup>13</sup>V. Dohm, University of Maryland Physics and Astronomy Department Technical Report No. 79-022 (unpublished), and to be published.

<sup>14</sup>P. C. Hohenberg, E. D. Siggia, and B. I. Halperin, *Phys. Rev. B* **14**, 2865 (1976).

<sup>15</sup>E. D. Siggia, *Phys. Rev. B* **13**, 3218 (1976).

<sup>16</sup>J. A. Tyson, *Phys. Rev. Lett.* **21**, 1235 (1968).

<sup>17</sup>We neglected here the frequency dependence of the order-parameter rate. The validity of this approximation will be discussed elsewhere.

## Spontaneous Polarization of Ferroelectric Triglycine Sulfate between 2.2 and 20 K

Sebastian Vieira

*Departamento de Física Fundamental, Universidad Autónoma de Madrid, Madrid, Spain*

and

Carmen de las Heras and Julio A. Gonzalo

*Departamento de Óptica y Estructura de la Materia, Universidad Autónoma de Madrid, Madrid, Spain*

(Received 11 September 1978)

The temperature dependence of the spontaneous polarization of ferroelectric triglycine sulfate has been determined in the range 2.2–20 K by measuring the charge released by a large crystalline sample with a Keithley electrometer. Above ~8 K this temperature dependence is  $\Delta P_s = Ce^{-\Delta/kT}$ , corresponding to a two-level system with energy splitting,  $\Delta = h\nu_E = 9.08 \times 10^{-15}$  erg, while between 2.2 and 4 K it is  $\Delta P_s = AT^{3/2}$ . Both temperature dependences are consistent with recent specific-heat measurements by Lawless.

Low-temperature specific-heat measurements<sup>1,2</sup> on a number of hydrogen-bonded and displacive ferroelectrics have consistently shown a contribution proportional to  $T^{3/2}$ . This contribution will eventually become dominant over the Debye  $T^3$  contribution as  $T \rightarrow 0$ . On the other hand, it has been suggested,<sup>3</sup> on thermodynamic grounds, that in zero field the specific heat and the spontaneous polarization (as  $T \rightarrow 0$ ) should both be proportional to the same power of  $T$ ; in particular  $C_0(T) = B'T^3$  would require  $\Delta P_s(T) = A'T^3$  (i.e.,  $\pi_1 = \partial P_s / \partial P_s / \partial T \propto T^2$ ), and in analogy with the ferromagnetic case,  $C_0(T) = BT^{3/2}$  would require  $\Delta P_s(T) = AT^{3/2}$ . These considerations imply that accurate measurements of possible slight changes in the spontaneous polarization as  $T \rightarrow 0$  would lend further support to the specific-heat findings of Lawless<sup>1,2</sup> if the same  $T$  dependence is observed. To our knowledge, no measurements of  $P_s$  down to 2.2 K have been reported in the literature. Thus, we think that the observations presented here are the first of their kind in any ferroelectric material. They appear to be

consistent with the specific-heat observations.

The samples were single-crystal plates, ranging in size from 5 cm<sup>2</sup> × 0.5 cm to 1 cm<sup>2</sup> × 0.2 cm, made available to us by B. Jimenez from the Consejo Superior de Investigaciones Científicas, Madrid. Gold electrodes were attached to the main surfaces (perpendicular to the ferroelectric  $b$  axis) by evaporation in a vacuum.

The samples were placed within the can of a liquid-He cryostat in which He gas at a pressure of 1 Torr was kept, to insure good thermal equilibrium. The larger sample, with which most measurements below 4 K were performed, was sandwiched between two stainless-steel plates (which provided for the electrical contacts) and freely suspended within the can to avoid inhomogeneous stresses.

This can was temperature controlled by means of an electronic controller. The sample temperature was measured by a calibrated Cryocal germanium resistor (using the standard four-terminal potentiometric method) attached to the ground electrode of the sample. Another thermometer