a carrier temperature of 10°K and increasing density  $n_0$  by a few percent. From our results we estimate that if EHL were created close to 0°K, its density would be  $(6.5-7) \times 10^{18}$  cm<sup>-3</sup>.

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## Determination of Relaxation Time in Bulk Superconductors

J. R. Leibowitz and M. C. Wilt

Department of Physics, Catholic University of America, Washington, D. C. 20064 (Received 4 February 1977)

A method for studying nonequilibrium effects and their  $\bar{k}$ -space anisotropy in bulk superconductors near  $T_c$  is reported. The relaxation time in single-crystal In determined directly by this method,  $\tau(t) = 0.77 \times 10^{-10} (1-t)^{-1/2}$ , represents confirmation, both in form and magnitude, of the Schmid-Schön predictions for the relaxation of the magnitude of the order parameter near  $T_c$ . The associated electron-phonon inelastic scattering time is in excellent agreement with that determined for the comparable portion of  $\bar{k}$  space by cyclotron resonance.

We report<sup>1</sup> a method for studying nonequillibrium superconductivity and its  $\bar{k}$ -space anisotropy in bulk superconductors near  $T_c$ , and the direct observation by this method of the relaxation time  $\tau(t)$  for high-purity, single-crystal indium satisfying  $\tau(t) = 0.77 \times 10^{-10} (1-t)^{-1/2}$  sec; here t is the reduced temperature, referred to the zero-field critical temperature  $T_c(0)$ . In thin-film experiments, Peters and Meissner<sup>2</sup> (Sn) and, later, Schuller and Gray<sup>3</sup> (Al) reported a divergence of  $\tau(t)$  near  $T_c$ . The data were found to be incompatible with the form  $(1-t)^{-1}$ , obtained<sup>4</sup> for gapless superconductors. It was inferred that the divergence must be of the form  $(1-t)^{1/2}$ .<sup>5</sup> It was possible in the present work to test the magnitude and form of the divergence of  $\tau(t)$  near  $T_c$  more definitively than heretofore, in part as a result of the following: (1) The  $T_{c}(0)$  broadening  $\delta T_{c}(0)$ <1 mK, as compared with 15-20 mK commonly encountered in thin films,<sup>6</sup> so that the form of the divergence could be meaningfully tested to t= 0.999; (2) the method minimizes commonly encountered complications due to subsequent pairbreaking (cf. Ref. 6) by a phonon emitted upon pair formation, and to quasiparticle branch imbalance<sup>7</sup>; and (3) the interaction has a  $\bar{k}$ -space

selectivity in the single crystal. The measured  $\tau(t)$  represents confirmation of the  $\Delta^{-1}$  temperature dependence, i.e., the form  $(1-t)^{-1/2}$ , predicted by Schmid and Schön<sup>8</sup> for the relaxation near  $T_c$  of the magnitude of the order parameter  $\Delta: \tau_{\Delta} = \pi^{3}T/7\zeta(3)\tau_{e-ph}\Delta^{-1}, \text{ where } \zeta \text{ is the Rie-mann zeta function and } \tau_{e-ph} \text{ the inelastic electron}$ scattering time by phonons. From our experimentally determined numerical coefficient 0.77 × 10<sup>-10</sup> (sec) we obtain  $\tau_{e-ph} = 1.2\overline{6} \times 10^{-10}$  (sec). [A factor of 2 is required by the fact that two quasiparticles are involved in each pair-forming process (cf., Ref. 5, p. 4869)]. This is in excellent agreement with the  $\tau_{e-ph}$  from (N state) cyclotron resonance,<sup>9</sup>  $\tau_{e-ph} = 1.27 \times 10^{-10}$  sec, identifiable with the same  $\bar{k}$ -space locus as is selected by the present interaction, i.e., the "cap" portion of the central-plane effective zone<sup>10</sup>  $\perp$  [001]. Note that  $\tau_{e-ph}$  is in general highly anisotropic, rendering questionable any detailed comparisons of  $\tau_{e-ph}(\vec{k})$  measured in polycrystals. Since the present method makes possible measurements on bulk single crystals and incorporates a k-space selectivity, it becomes possible to identify measured times  $\tau_{\Delta}$  and  $\tau_{e-ph}$  with particular regions of Fermi surface, and directly to determine



FIG. 1. Attenuation vs temperature, as a function of applied field  $H_A$ . Inset: configuration of N-S laminas relative to  $\bar{\mathbf{q}}$ ,  $\bar{\boldsymbol{\epsilon}}$ , and  $\bar{\mathbf{H}}_A$ .

their  $\bar{k}$ -space anisotropy. The latter effort is currently underway.

The method uses ultrasonic shear waves (injected transverse phonons) to impose a rapid, oscillatory displacement near  $T_c$  of normal-superconducting (N-S) interfaces, as exist for example in the alternating N and S laminas of the intermediate state. The method is, in principle, applicable to other type I superconductors, and possibly to low-K, bulk type II as well. The configuration of wave propagation and polarization vectors  $\vec{q}$  and  $\vec{\epsilon}$ , applied field  $\vec{H}_A$ , and N and S laminas in the sample (diameter  $\approx 1$  cm, thickness  $\approx 1$  mm) are shown schematically in Fig. 1 (inset);  $H_A$  is inclined at angle  $\beta$  relative to surface normal in order to promote ordering<sup>11</sup> of laminas. In the present experiments wave frequency range is f = 75 - 195 MHz, the t range is  $t \approx 0.99$  to t > 1, and field range is  $0 \leq H < 2.0$  G. Under these conditions  $ql \gg 1$  and the transversephonon-quasiparticle interaction is dominated<sup>10, 12, 13</sup> by electromagnetic coupling, so that there is generated a local induction field  $\delta H$  because of the time-varying local ionic and electronic currents identified with the transversephonon-electron coupling; here l is the electron mean-free path.

How does the electromagnetic coupling of the transverse-phonon-electron interaction drive the N-S interface oscillation? In the intermediate state and for  $\vec{H}_A$  normal to the sample surface,<sup>11</sup>  $d_N/d \approx H_A/H_C(T)$ , so that  $\delta d_N/d \approx \delta H/H_C(T)$ ; here  $d_N/d$  is the relative width of N laminas  $[d = d_N + d_S]$ , and  $\delta d_N$  is the modulation of the N laminar

width. That is, a modulation  $\delta d_N$  of normal laminar thickness is imposed by field modulation  $\delta H$ ; δH introduces a high-frequency "breathing mode" in the N-S laminas, the motion dictated by the intermediate state constraint that  $H = H_C$  in the N domains. The N-S interface is, of course, not to be viewed as a conventional mechanical boundary (e.g., phonon velocities are the same in N and S regions to 1 part in  $10^6$ ). Further, the field changes associated with the breathing mode do not involve transport of flux associated with an externally applied field. Rather, the field  $\delta H$ originates in the time varying local currents associated with the electron-phonon coupling. Since the time variation is imposed not by the external field  $H_A$ —which variation would be strongly restricted by screening currents—but rather by the propagating transverse-phonon-electron coupling and its associated time-varying local induction field,  $\delta H$  responds with the impressed wave frequency  $\omega$ . It is for this reason that the "breathing mode" in the N volume can be established.

In the given T range, the accessible shear wave amplitudes and frequencies are of sufficient magnitude to make the local field  $\delta H$  competitive with the critical field  $H_c(T)$ . The effect is that a significant portion of the total sample volume, a fraction proportional (at given frequency) to wave amplitude, can be affected. It is therefore anticipated that there will appear, near  $T_c$  and at sufficiently high frequencies, a discernible relaxational absorption contribution  $\delta \alpha$  associated with the modulated volume  $\delta V$  superposed on the "baseline" attenuation contributions already reported.<sup>10, 12-16</sup> While the interaction is taken to be linear, the thickness  $\delta d_{\mathrm{N}}$  (and hence the volume  $\delta V$ ) will increase with wave amplitude A and frequency  $\omega$  (both of which increase the amplitude of  $\delta H$ ) and the relaxational absorption contribution, accordingly, will become larger. Accordingly, in our procedure for determining  $\tau(t)$ , amplitude A and frequency f are kept fixed (as will be seen later).

Such a relaxational absorption model as the one presented here was proposed in 1968 by Leibowitz<sup>17</sup> for the absorption peak observed<sup>18</sup> near  $T_c$  and under small applied field  $H_A$  in indium by Leibowitz and Fossheim. While the proposed model<sup>17</sup> anticipated identification of the peak with relaxation of the order parameter, technical limitations at that time prevented a quantitative test. But in the present work this mechanism is, in point of fact, clearly indicated, and a method found for making quantitative determination near  $T_c$  of the effective relaxation time  $\tau$  and its T dependence.

In Fig. 1 the dependence on T of wave attenuation  $\alpha$  is shown near  $T_c$  for given f and A values. In these data the approach to the transition is made from the N state. Complete reversibility of  $\alpha(t)$  is then observed so long as T does not fall below the intermediate state regime.] Note that for given H the attenuation rises at  $T_{c}(H)$ , before falling with further reduction of T: It is to be emphasized that a peak which can cause the total attenuation to exceed the N state level [the attenuation for  $T > T_c(H)$ ] and which is amplitudedependent, as noted earlier, is incompatible with the steady-state attenuation contributions already reported in the literature (see Refs. 10, 12-16). Also, since  $H \leq 2$  G there are no relevant magnetoacoustic contributions; and, since  $\hbar\omega \ll \Delta(T)$ , except within ~ $10^{-5}$  K of  $T_c$ , no single ultrasonic phonon pair-breaking effects enter. Finally, the attenuation peak near  $T_c$  (Fig. 1) is readily separable from the attenuation structure observed by Lin and Leibowitz<sup>16</sup> and identified with an equilibrium spatial variation of the order parameter: On fundamental grounds the latter mechanism relies on sharply defined boundary conditions, which should be removed at large wave amplitudes—precisely the condition required for the effects of interest in the present paper. Further, the reference temperature in the "superlattice" effect reported by Lin and Leibowitz<sup>16</sup> is required to be  $T_c(H)$ , while that for the effects of interest in the present paper is  $T_c(0)$ . As will be detailed elsewhere, the distinctions represented by these criteria have been demonstrated experimentally: The superlattice structure disappears at high amplitudes, where the relaxational peak becomes important; the distinct roles of the two reference temperatures are also observed.

There is an annoying obstacle to demonstrating the peak is identifiable with a *T*-dependent relaxation time  $\tau(T)$  [and that, for a given  $\omega = 2\pi f$ ,  $\tau$ reaches a maximum value at temperature *T* for which  $\tau(T) \approx 1/\omega$ ]. A competing effect cuts off the low-temperature side of the absorption peak: Since  $H_c(T)$  is strongly *T* dependent near the phase transition  $(\Delta H/\Delta T \approx 0.2 \text{ G/mK})$  and since  $\delta d_N/d \approx \delta H/H_c(T)$ , the rapid increase of  $H_c(T)$ with the decrease of *T* is responsible for a corresponding reduction in volume occupied by the breathing mode. Hence, the peak is cut off on its low-*T* side, and its observed maximum does not reflect the true  $\tau(t)$  form. To address this problem, it will not suffice on technical grounds to find the  $\tau(T_i)$  values as follows: For each fixed value of  $H_A$  find f corresponding to  $\delta\alpha$  (max) and from the corresponding  $T_i$  location of  $\delta \alpha$  (max), together with the wave frequency  $\omega$ , obtain a value for  $\tau(T_i)$ . Then repeat the process for another  $H_A$  value, yielding another  $T_j$  and  $\tau(T_j)$ , and so on. A problem with this procedure is that it is not possible, especially at high rf frequencies, to be certain that when the transmitted frequency is changed, the amplitude of particle displacement at the first sample interface is not also altered. (It is, of course, not sufficient that the measured peak voltage out of the transmitter or even at the site of the transmitting transducer, be maintained constant.) Other procedures for extracting  $\tau(T)$  suggest themselves. While we cannot pursue the problems in detail here, it should be noted that methods which introduce wave amplitude A as a parameter, either intentionally or by inadvertence, contain difficulties. For example, the shifts of  $\alpha(T)$  with A cannot be attributed entirely to the effect<sup>15</sup> of local field  $\delta H$ , since the observed shift can also include sample self-heating relative to the thermometer.

The device adopted in the present work, therefore, was to use  $H_A$  as a parameter in generating, for fixed frequency f and wave amplitude A, families of curves of  $\alpha(T)$  and subtracting the baseline attenuation discussed earlier. A few such  $\alpha(T)$  curves are illustrated in Fig. 1 (arbitrary relative dB offset). In this way  $\tau(T)$ , referred to  $T_c(0)$ , could be reconstructed for the observed peaks near  $T_c(H)$ , as  $T_c(H)$  was increasingly shifted to lower temperatures from  $T_c(0)$ by stepwise increase of  $H_A$ . The result of this effort for the particular frequencies 135 and 165 MHz is shown in Fig. 2 in which the data points



FIG. 2. Maximum peak height vs temperature, identifying  $\omega_i \tau_i(t_i)$  for each frequency  $f_i$ .



FIG. 3. Temperature dependence of relaxation time  $\tau \approx 1/\omega$ .

refer, for each frequency, to  $\delta \alpha(T)$  values derived from data such as indicated in Fig. 1, after baseline subtraction. At each frequency the maximum  $\delta \alpha(T)$ , for which  $\omega \tau \approx 1$  in Fig. 2, identifies that T for which  $\omega \tau \approx 1$ . Note, that in Fig. 2 the absorption peak for the 165 MHz case reaches a maximum about 16 mK from  $T_c(0)$ , while the width of the peaks in the curves of Fig. 1 is only about 5 mK. From such data such as are illustrated in Fig. 2, a value for  $\tau(T) \approx 1/\omega$  is determined for each frequency. No fitting parameters, such as "curve fitting" shifts of  $T_c$ , are introduced. The results are summarized in Fig. 3. Note that in both Figs. 3(a) and 3(b) the curves are constrained by intercept as well as straightline fit, which fixes the point of intersection with ordinate in Fig. 3(a) and the crossover point of the curves in Fig. 3(b). In Fig. 3(b) the temperature dependence  $\tau(T) \sim (1-t)^n$  is examined for n = -1 and  $n = -\frac{1}{2}$ .

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