

spin-lattice interaction, and (b) a single phonon with energy  $2h\nu$  produced by the anharmonic interaction of the lattice is absorbed by means of the ordinary one-phonon spin-lattice process. The latter possibility is rejected in this case because the detection system, which has a narrow bandwidth about  $\nu$ , will not respond directly to a frequency of  $2\nu$ . Other, higher order processes involving second-harmonic phonons are rejected because of low probability and because effects similar to those reported here have not so far been observed for other paramagnetic ions in  $\text{CaF}_2$ .<sup>12</sup> Furthermore, a one-parameter fit of Eq. (5) to be experimental points (shown in Fig. 2 as a curve) is quite good.

In conclusion, we have observed the attenuation of elastic waves due to direct spin-lattice interactions involving multiple-phonon absorption. Quantitative measurements of the angular variation of the resonant magnetic field and the UPR attenuation as a function of ultrasonic intensity have been explained by assuming that two phonons are (essentially) simultaneously annihilated and the population of an excited spin state increased by unity. We have also confirmed that non-Kramers ions can be strongly coupled to the lattice.

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## DIAMAGNETIC AND PARAMAGNETIC SURFACE CURRENTS INDUCED BY TEMPERATURE CHANGES IN TYPE-II SUPERCONDUCTORS IN STATIC MAGNETIC FIELDS\*

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We present evidence that a change in surface currents from critical paramagnetic to critical diamagnetic (or vice versa) is induced by a suitable increase (or decrease) of the temperature of a semireversible type-II superconductor immersed in a static magnetic field  $H < H_{c2}$ .

Recent work has shown that the surface layer of type-II superconductors in the mixed state can support net macroscopic persistent currents.<sup>1-4</sup> This current is termed diamagnetic (paramagnetic) when the magnetic field it produces inside the specimen opposes (aids) the applied field. These surface currents play

an important role in the hysteretic behavior of type-II superconductors.<sup>3</sup> A change in the surface current from critical diamagnetic to critical paramagnetic (or vice versa) can be induced by a suitable decrease (or increase) of the external magnetic field at constant temperature. In this Letter we present evidence

that a change in the surface current from critical diamagnetic to critical paramagnetic (or vice versa) can be induced by a suitable decrease (or increase) of the temperature in a static magnetic field.

We present data for a solid cylinder of high-purity  $\text{Pb}_{0.84}\text{In}_{0.16}$  of  $\approx 10$  cm length, 0.25 cm diam in a magnetic field parallel to the axis of the cylinder. The magnetization is monitored by electronically integrating the emf from a pick-up coil placed around the sample.<sup>5</sup> A non-inductive single-layer coil of Manganin wire (0.0075 cm diam) close wound directly on the sample enables us to control the temperature of the sample.

Figure 1 shows the magnetic behavior of the sample at 4.2°K in an increasing and a decreasing magnetic field and the partial Meissner effect after cooling through  $T_c$  to 4.2°K in various static magnetic fields (denoted  $H_{\text{cool}}$ ). The solid lines in this figure show that, typically, the locus of the magnetization, upon reversal of the magnetic field sweep, traces paths with slope  $\Delta M/\Delta H = -1/4\pi$  over a range of field change. The slopes begin to deviate from  $-1/4\pi$  at  $H_f'$  and  $H_f''$ . Since  $4\pi\langle M \rangle = \langle B \rangle - H$ , it is clear that  $\langle B \rangle$  is constant as  $H$  varies from  $H_r'$  to  $H_f'$  and  $H_r''$  to  $H_f''$ . Applying Faraday's and Lenz's law of induction to the

behavior when the field sweep is reversed, we deduce that a critical diamagnetic (paramagnetic) current is quenched and a critical paramagnetic (diamagnetic) current is induced at the surface of the sample as the external field proceeds from  $H_r'$  to  $H_f'$  ( $H_r''$  to  $H_f''$ ). This conclusion is substantiated by the following observations<sup>3</sup>: When the sample is coated with an antiferromagnetic or high-conductivity metal, (i) the locus of the magnetization above  $H_{c1}$  in increasing (decreasing)  $H$  lies below (above) the locus of Fig. 1, and (ii) the Meissner effect is enhanced. We note (see Fig. 1) that  $|H_f' - H_r'| = |H_f'' - H_r''|$  within experimental accuracy when  $\frac{1}{2}(H_f' + H_r') = \frac{1}{2}(H_f'' + H_r'')$  whether  $H$  decreases or increases upon reversal (provided that  $H$  has previously exceeded  $H_{c1}$ ).

In Fig. 2 we again present the partial Meissner effect at 4.2°K vs  $H_{\text{cool}}$  and show the locus of the magnetization upon an increase or a decrease of the magnetic field from  $H_{\text{cool}}$  when (a)  $0 < H_{\text{cool}} \lesssim H_{c1}$  and when (b)  $H_{c1} < H_{\text{cool}} < H_{c2}$ . We note the similarity between the behavior of the locus of the magnetization starting at  $D$  of Fig. 2(b) and  $B$  of Fig. 1. In both cases (i)  $\Delta M/\Delta H = -1/4\pi$  as  $H$  increases from  $H_{\text{cool}}$  or  $H_r''$  to  $H_f''$  and (ii)  $|\Delta M/\Delta H| < 1/4\pi$  as  $H$  decreases from  $H_{\text{cool}}$  or  $H_r''$ . Further

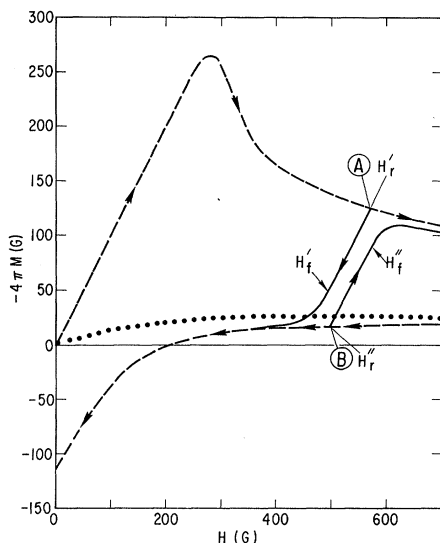


FIG. 1.  $T = 4.2^\circ\text{K}$ . Dashed curves: magnetization in field increasing from 0 and decreasing from  $H \approx H_{c2}$ . Solid curves: typical locus of the magnetization upon reversal of field sweep. Dots: data points for Meissner effect diamagnetism versus static field present during cooling.

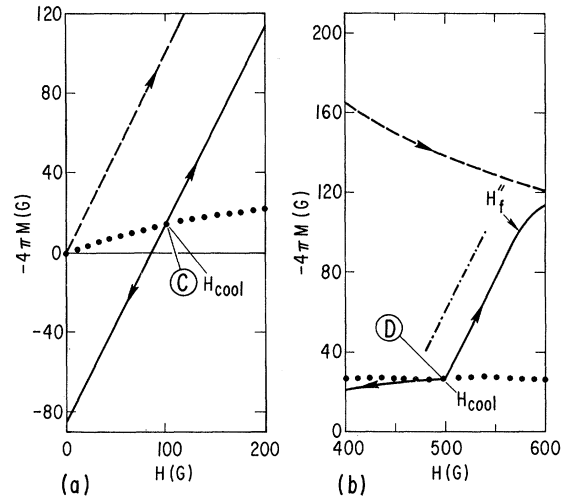


FIG. 2. Dashed curves: portions of initial magnetization at 4.2°K. Dash-dot curve:  $\Delta M/\Delta H = -1/4\pi$ . Dots: data points for Meissner effect versus static field present during cooling. Solid curves: typical magnetic response of sample to an increase or a decrease of applied field after cooling through  $T_c$  to 4.2°K in a static magnetic field, denoted  $H_{\text{cool}}$ , where (a)  $H_{\text{cool}} = 100 \text{ G} < H_{c1}$  and (b)  $H_{\text{cool}} = 500 \text{ G} > H_{c1}$  ( $H_{c1} \approx 250 \text{ G}$  at 4.2°K).

we note that  $H_f'' - H_{cool} = H_f'' - H_r''$  when  $\frac{1}{2}(H_{cool} + H_f'') = \frac{1}{2}(H_r'' + H_f'')$ . We conclude that the surface carries a critical paramagnetic current after cooling through  $T_c$  to 4.2°K in a static magnetic field  $H_{c1} \lesssim H_{cool} < H_{c2}$ .<sup>4</sup> This critical paramagnetic current is quenched and a critical diamagnetic current is induced at the surface as  $H$  increases from  $H_{cool}$  to  $H_f''$ .

In Fig. 2(a), an initial slope  $\Delta M/\Delta H = -1/4\pi$  is encountered whether  $H$  is increased or decreased from  $H_{cool}$ , indicating that the net surface current  $I_s$  is subcritical before  $H$  is varied when  $0 < H_{cool} \lesssim H_{c1}$ . Presumably  $I_s = 0$  after cooling through  $T_c$  with  $H = 0$ . Since  $I_s$  is critical paramagnetic when  $H_{c1} \lesssim H_{cool} < H_{c2}$  we may infer that  $I_s$  varies from 0 to critical paramagnetic as  $H_{cool}$  spans the range from 0 to  $\gtrsim H_{c1}$ . This conclusion is consistent with all our observations on these samples.

Figure 3 shows typical hysteretic behavior of the partial Meissner effect as the temperature is cycled from  $T_c$  to 4.2°K to  $T_c$  and varied over intermediate ranges of temperature in a static magnetic field.<sup>6</sup> The curves are reproducible within the thickness of the lines

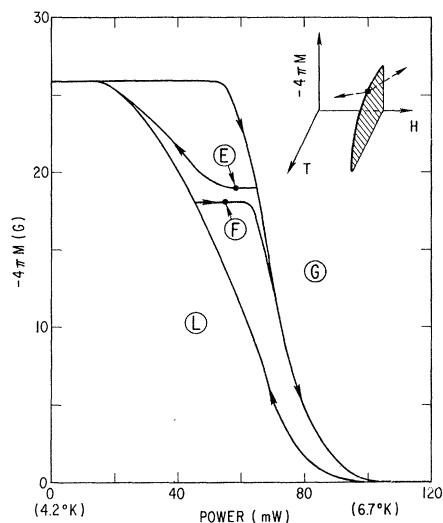


FIG. 3. Curve L (G) shows increase (decrease) of Meissner-effect diamagnetism as  $T$  is decreased from  $T_c$  to 4.2°K (increased from 4.2°K to  $T_c$ ) in a static magnetic field  $H = 600$  G. Temperature varies monotonically with power dissipated in heater. Intermediate curves show locus of magnetization as temperature sweep is reversed. Inset shows schematically paths in  $M-H-T$  space as temperature sweep in a static field is interrupted and applied field  $H$  is subsequently increased or decreased at constant  $T$  (heater power). Locus of magnetization is continuously monitored during changes of  $T$  and  $H$ .

drawn in Fig. 3 when  $\Delta T/\Delta t \approx 1^\circ\text{K}/\text{min}$ . The hysteretic behavior of  $4\pi M$  vs  $T$  at constant  $H$  and, in particular, the observation that curve G lies above curve L in Fig. 3 can be qualitatively understood in terms of (i) pinning forces due to imperfections and impurities, and/or (ii) a multiply connected surface layer capable of supporting net paramagnetic or diamagnetic currents. Clearly, both (i) and (ii) will oppose exit of flux along curve L and also oppose entry of flux along curve G. Exit or entry of flux indicates that these opposing "forces" are being overcome. The concepts exploited to account for the familiar hysteresis in the  $4\pi M-H$  plane at constant  $T$  are also applicable to hysteresis in the  $4\pi M-T$  plane at constant  $H$ .

Starting from  $T > T_c$  for every measurement and interrupting the "temperature sweep" at various points in the  $4\pi M-T$  plane, we have explored the magnetic response of the sample to an increase or a decrease of the magnetic field from the chosen static value as shown schematically in the inset of Fig. 3. The behavior at  $T = 4.2^\circ\text{K}$  when  $H_{c1} \lesssim H_{cool} < H_{c2}$  was presented in Fig. 2.

The response exemplified by Fig. 2(b) is encountered on the decreasing  $T$  and ascending  $|-4\pi M|$  portion of curve L of Fig. 3. We conclude that the surface layer carries a critical paramagnetic current on this portion of curve L before  $H$  is varied. Figure 4(a) presents

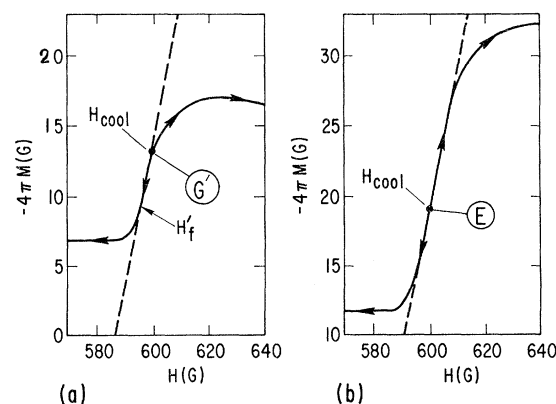


FIG. 4. Dashed lines:  $\Delta M/\Delta H = -1/4\pi$ . Solid curves: typical magnetic response of sample at constant  $T$  (heater power) to an increase or a decrease of the applied field from  $H_{cool}$ , the field present during the temperature variations. In (a) increasing temperature sweep is interrupted at an arbitrary point on the descending part of curve G of Fig. 3, and in (b) temperature sweep is interrupted at E of Fig. 3.

the typical response encountered on the increasing  $T$  and descending  $|-4\pi M|$  portion of curve  $G$ . We note the equivalence of the behavior starting at  $G'$  of Fig. 4(a) and that starting at  $A$  of Fig. 1. In both cases (i)  $\Delta M/\Delta H = -1/4\pi$  as  $H$  decreases from the existing field  $H_{\text{cool}}$  or  $H_{\gamma'}$  to  $H_f'$ , and (ii)  $|\Delta M/\Delta H| < 1/4\pi$  as  $H$  increases from the existing field  $H_{\text{cool}}$  or  $H_{\gamma'}$ . We conclude that the surface layer carries a critical diamagnetic current on this portion of curve  $G$  before  $H$  is varied. Both these conclusions seem inescapable if (a) we acknowledge the existence of a multiply connected surface layer capable of supporting net diamagnetic or paramagnetic currents, and (b) consider the fact that on the portions of curves  $L$  and  $G$  under discussion, the Meissner-effect diamagnetism is increasing (decreasing), i.e., flux is leaving (entering) the specimen as  $T$  is decreasing (increasing) with a static field present.

Figure 4(b) presents the typical response encountered in the vicinity of the midway point of the traverse between the ascending and descending parts of curves  $L$  and  $G$ ; viz.,  $E$  and  $F$  of Fig. 3. An initial slope  $\Delta M/\Delta H = -1/4\pi$  is encountered whether  $H$  is increased or decreased from  $H_{\text{cool}}$  indicating that  $I_c$  is subcritical before  $H$  is varied. The magnetic response of the specimen changes through the sequence (exemplified by) Figs. 2(b), 4(b), and 4(a) (and vice versa) as  $T$  is varied between the ascending and descending  $|-4\pi M|$  curves (and vice versa). Clearly then, the surface current varies from critical paramagnetic to subcritical to critical diamagnetic (and vice versa) as  $T$  is varied over suitable intervals in the presence of a static field. Hence, a decrease (increase) in  $T$  at constant  $H$  is analogous to a decrease (increase) in  $H$  at constant  $T$ .

Finally we note that  $4\pi M$ , hence  $\langle B \rangle$ , remains constant over an appreciable fraction of the traverse between the ascending and descending  $|-4\pi M|$  curves in Fig. 3. Since the surface current is changing in magnitude and sense during the corresponding "temperature sweep," it is clear that an appreciable redistribution of flux also must occur inside the specimen in order that  $\langle B \rangle$  remain constant.

In the presence of a constant field, as  $T$  decreases below  $T_c$ , migration of flux filaments towards the surface presumably occurs when the outward forces they experience exceed the

inward forces and the pinning strength of imperfections in the bulk. The laws of induction and the existence of a multiply connected surface layer capable of supporting net macroscopic currents require that a paramagnetic current arise in this surface layer to oppose exit of flux. This surface current will grow until it "saturates" or until the outward forces experienced by the flux filaments are balanced. The occurrence of a Meissner-effect diamagnetism indicates that this balance occurs after the surface current has attained a critical state and "excess" flux has been expelled. Once a balance has been achieved, the surface current may become subcritical if the current-carrying capacity of the surface layer increases faster than required to maintain  $\langle B \rangle$  constant as  $T$  is lowered further. Our results suggest that the latter situation is encountered when  $H_{\text{cool}}$  becomes  $\lesssim H_{c1}$  as  $T$  decreases [see Fig. 2(a)]. Our conclusion that a critical or subcritical paramagnetic surface current vanishes and a critical diamagnetic current is generated as  $T$  increases in a static field combined with Maxwell's equation  $dB_z/dr = -4\pi j_\theta/10$  requires that an inward migration of flux filaments away from the surface takes place. This in turn implies changes in the configuration of currents in the bulk of the specimen. According to the model we have presented to account for our observations, slow oscillations of  $T$  of suitable amplitude are accompanied by oscillations in the magnitude and sense of macroscopic currents in the surface layer and in the bulk of a semireversible type-II superconductor in a stationary magnetic field. Investigation of  $B$  versus radial position<sup>7</sup> as  $T$  is varied at constant  $H$  should yield interesting data on the mutual interactions of flux filaments and their interaction with surface currents.<sup>8</sup>

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## TRANSVERSE ELECTROREFLECTANCE

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New techniques for observing the Seraphin effect are presented, which offer additional symmetry information on optical interband thresholds, and eliminate photon-energy restrictions inherent in previous techniques. Examples on GaAs and ZnSe are shown.

Previous electroreflectance studies<sup>1-7</sup> have been restricted to semiconductor materials in which sufficient conductivity exists to transfer charge carriers into and out of the surface barrier region [surface-barrier electroreflectance (SBE) or the Seraphin effect]. These experiments have yielded a wealth of precise new or confirming information on interband threshold energies.<sup>8</sup> However, studies of transition energies above 5.5 or 6 eV, and interpretation of line-shape data, have been handicapped by the inhomogeneity of the modulating electric field, the necessity of highly-doped semiconductor materials, and the wavelength limitations inherent in the transparent electrode. These handicaps have now been obviated by development and successful application of a most straightforward method of electric field application to the reflecting sample (see Fig. 1). The result is a transverse electroreflectance (TE) technique applicable to materials of about  $10^8 \Omega \text{ cm}$  or greater resistivity.

This new technique offers the following advantages: (1) Because the electric field is applied transversely to the light propagation direction, no optical window or transparent electrode medium is required in front of the reflecting surface, thereby eliminating restrictions on the optical energy range available for study. (2) With polarized incident light, experiments may be performed with the optical elec-

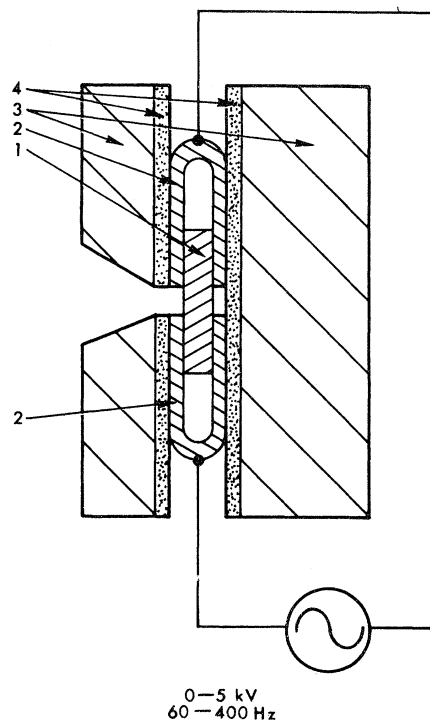


FIG. 1. Schematic diagram of the experimental arrangement for transverse electroreflectance. The sample is labeled 1, the Be-Cu electrodes (operated balanced to ground) are labeled 2, the ground-potential cooling finger is labeled 3, and the Teflon insulation (0.010 in. thick) is labeled 4. The electronic signal processing is similar to that described by Seraphin (Ref. 3).