INHERENT LOW-FREQUENCY LOSSES OF THE SUPERCONDUCTING SURFACE SHEATH*

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Strongin et al.[1] have measured at 18 cps the real (χ_1') and imaginary (χ_1'') parts of the magnetic susceptibility and have found a power loss ($\chi_1''>0$) for a cylinder of Pb-0.1% Bi (type-I superconductor with $\kappa>0.417$) when the static field was between the thermodynamic critical field H_c and the surface nucleation field H_{c3}. This means that the surface sheath produces ac losses. χ_1'' has a peak as a function of the applied (static) magnetic field H_0 and it is remarkable that $\chi_{1\,\max}''$ is larger than the imaginary part of the normal state susceptibility χ_n''. Furthermore, Sandiford and Schweitzer[2] and also Barnes and Fink[3] have shown that hysteresis loops of the ac and dc magnetization as a function of H_0 can be swept out in the superconducting sheath state. This is also equivalent to a power loss.

We shall show that these losses are an inherent feature of the critical state[4] of the surface sheath and that flux pinning is not required to account for the low-frequency losses. (Note that the flux in the superconducting surface sheath is not quantized in the same sense as in the mixed state.) We shall calculate χ_1' and χ_1'' for the superconducting surface sheath of a long macroscopic cylinder of radius R with the static magnetic field H_0 parallel to its axis and with an ac magnetic field $h=h_0\sin\omega t$ superimposed on H_0. We shall show that the peak of χ_1'' as a function of H_0 can be simply related to the critical-state properties of the surface sheath, and that $\chi_{1\,\max}''$ is of constant value regardless of material properties and is always larger than χ_n''. It is postulated that part of the ac losses between the lower critical field H_{c1} and the upper bulk critical field H_{c2} are due to the surface sheath surrounding the mixed state of the sample.

It has been shown[4] that the magnetization per unit volume of the surface sheath in the critical state is approximately

$$4\pi M_1 = \pm\gamma H_{c2} f, \qquad (1)$$

where $\gamma=\kappa^{-1}(\lambda/R)^{1/2}$, $H_{c2}=\sqrt{2}\,\kappa H_c$, and $f=(\Delta/\xi)\times[F^2(R)/\kappa]$ is a function of H_0/H_{c2} and κ and is tabulated in Ref. 4. As the sample is capable of shielding the interior of the cylinder over part of the magnetic field cycle[2,3] as shown for example in Fig. 1 between the points g and i and also between l and j, it is plausible to assume when one sweeps along these diamagnetic lines that the magnetic field near the surface in the superconducting sheath is approximately kept constant and of the same value as in the interior of the cylinder, so that to the next approximation Eq. (1) can be written when ϵ is small compared to unity as

$$4\pi M_1 = \mp\gamma H_{c2} f(1\pm\epsilon), \qquad (2)$$

where the upper signs apply to increasing magnetic fields and the lower signs to decreasing magnetic fields and $\epsilon=\gamma\,|df/d(H_0/H_{c2})|$. Eq. (1)

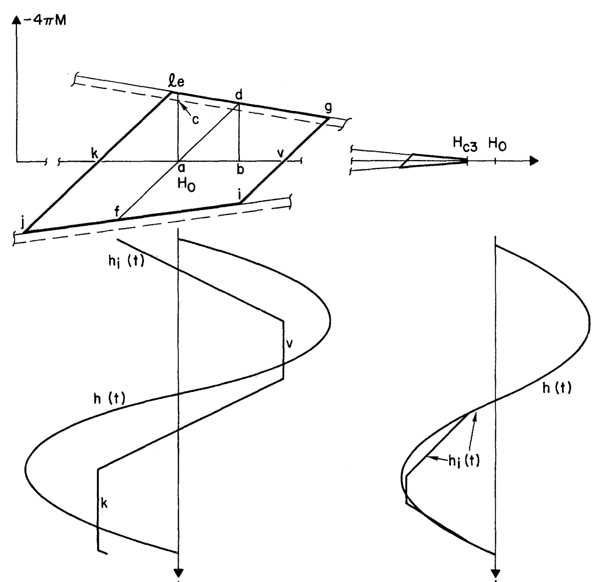

FIG. 1. Hysteresis loop of the magnetization (per unit volume) as a function of the applied magnetic field when the ac amplitude h_0 is larger than $4\pi M_1$, where $4\pi M_1$ is the critical magnetization per unit volume[4] of the surface sheath. Shown are also the time-dependent applied $[h(t)]$ and internal[6] $[h_i(t)]$ magnetic fields for a macroscopic cylinder. On the left-hand side the static magnetic field H_0 is between H_{c2} and H_{c3} (type-II super-superconductor) or H_c and H_{c3} (type-I superconductor), and on the right-hand side $H_{c3}<H_0<H_{c3}+h_0$.

corresponds to the dashed lines in Fig. 1 and Eq. (2) to the solid lines (e.g., l-g and j-i). The values of ϵ/γ are calculated from the data of Ref. 4 and are shown in Fig. 2. For the present consideration we shall assume that the sweep rate $dH_0/dt \ll \omega h_0$, $R \gg \lambda$ (low-field penetration depth), and also $R \gg \Delta$ (thickness of the surface sheath). ϵ is then always small compared to unity.

Assume that we have increased the magnetic field H_0 to that corresponding to point a in Fig. 1 and that the sheath is in the critical state at point e. Let us now apply $h(t)$ and increase its amplitude h_0 from zero to h_c (h_c is equal to the distance a-b, b-d, and also a-c). The time-averaged magnetization per unit volume $4\pi M$ (averaged over many cycles of ω) decreases then along the line e-a and vanishes[5] when $h_0 = h_c = \gamma H_{c2}|f|$. The time-dependent part of the magnetization varies always along a diamagnetic line for $h_0 \leq h_c$ when h_0 is kept constant. Therefore a cylinder with a surface sheath appears perfectly diamagnetic as far as the ac signal is concerned[1] ($\chi_1' = -1/4\pi$ and $\chi_1'' = 0$). When $h_0 > h_c$ a hysteresis loop is swept out such as that shown in Fig. 1—for example, $g \to i \to j \to l \to g$. For $h_0 > h_c$ the internal magnetic field as a function of time is then approximately that indicated[6] by $h_i(t)$ in Fig. 1, and its phase relation is compared to that of the applied ac field $h(t)$. Over the interval g-i, the internal field is kept constant at the value v, and over j-l it is kept constant at the value k.

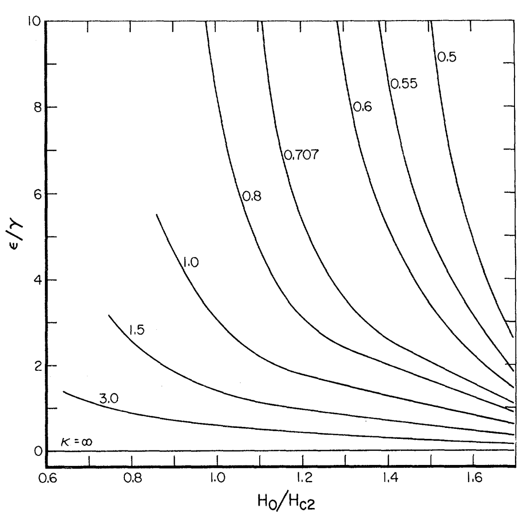

FIG. 2. The function $\epsilon/\gamma = |df/d(H_0/H_{c2})|$ is shown for various values of H_0/H_{c2} and κ, where $f = (\Delta/\xi) \times [F^2(R)/\kappa]$.

The ac susceptibilities are calculated the following way. The internal field is

$$H_i = H_a + 4\pi M = H_0 + h_i, \tag{3}$$

where the applied field $H_a = H_0 + h_0 \sin\omega t$ and

$$M = m_0 + h_0 \sum_{\alpha=1}^{\infty} (\chi_\alpha' \sin\alpha\omega t - \chi_\alpha'' \cos\alpha\omega t). \tag{4}$$

m_0 is an induced dc component of the magnetization which arises because of the asymmetry of $h_i(t)$ with respect to time. When $\epsilon \ll 1$ the function h_i becomes approximately symmetric and its Fourier series is

$$h_i = (h_0 - h_c)\left\{\epsilon + \frac{4}{\pi}\left[\left(\frac{\sin a}{a}\right)(\sin a \sin\omega t - \cos a \cos\omega t) - \frac{1}{3}\left(\frac{\sin 3a}{3a}\right)(\sin 3a \sin 3\omega t - \cos 3a \cos 3\omega t) + - \cdots\right]\right\}, \tag{5}$$

where $\cos a = (h_c/h_0)^{1/2}$. Experimentally, one observes voltages proportional to the ac component of M at one frequency only and this is usually the in-phase component of the fundamental of M which is proportional to χ_1', and the out-of-phase component of the fundamental of M which is proportional to χ_1''. From Eqs. (3) to (5), one obtains for the fundamental components for $h_0 \geq h_c$

$$\chi_1' = -\frac{1}{4\pi} + \frac{1}{\pi^2}\left(1 - \frac{h_c}{h_0}\right)^{3/2}\left(\frac{\sin a}{a}\right), \tag{6a}$$

$$\chi_1'' = \frac{1}{\pi^2}\left(1 - \frac{h_c}{h_0}\right)\left(\frac{h_c}{h_0}\right)^{1/2}\left(\frac{\sin a}{a}\right), \tag{6b}$$

and for the induced dc component

$$4\pi m_0 = \epsilon(h_0 - h_c). \tag{7}$$

Similar expressions are obtained for the higher odd-harmonic components. The even-harmonic components are zero within our approximation.

When H_0 is near H_{c3} the above calculations are only very approximate. A more accurate solution is obtained for $H_{c3} \leq H_0 \leq H_{c3} + h_0$ from the right-hand schematic in Fig. 1. For this calculation we assume for the time being that the susceptibilities in the normal state χ_n' and χ_n'' are zero. This is strictly correct only when $R \ll \delta$ (rf skin depth). We shall discuss this point in more detail below. When $H_0 = H_{c3}$ and terms of order ϵ and smaller are neglected (but not of order $\epsilon^{1/2}$), one obtains

$$\chi_1' = -\frac{1}{8\pi}\left[1-\frac{8}{\pi^2}\left(1+\frac{2}{\pi}\epsilon^{1/2}\right)\right], \tag{8a}$$

$$\chi_1'' = [(\pi-2)/\pi^4]\epsilon^{1/2}, \tag{8b}$$

and when H_0 is near $H_{c3}+h_0$, one gets

$$\chi_1' = -\frac{1}{4\pi}x^3[\tfrac{2}{3}-\tfrac{1}{2}(1+\tfrac{1}{2}\epsilon^{1/2})], \tag{9a}$$

$$\chi_1'' = \frac{1}{48\pi^2}x^4(2\epsilon)^{1/2}, \tag{9b}$$

where $x^2 = 2(H_{c3}+h_0-H_0)/h_0$.

Let us estimate the general shape of χ_1' and χ_1''. This estimate is in good agreement with the experiment.[1] When $h_c/h_0 \leq 1$, we obtain $\chi_1' = -1/4\pi$ and $\chi_1'' = 0$. When $h_c/h_0 > 1$ the absolute value of $|\chi_1'|$ decreases while χ_1'' increases. Both functions have horizontal tangents as a function of H_0 at $h_c/h_0 = 1$. The static magnetic field H_0 which satisfies the condition $h_c = h_0$ depends on κ, R, λ, H_c, and h_0. This magnetic field can be anywhere between H_{c2} and H_{c3} (type-II superconductor) or H_c and H_{c3} (type-I superconductor). While $|\chi_1'|$ drops smoothly to a small value at $H_0 = H_{c3}$ and to zero at $H_0 = H_{c3}+h_0$, χ_1'' reaches a maximum at $h_c/h_0 = 0.385(2)$ whose value is $\chi_{1\,\max}'' = 0.334/\pi^2$. χ_1'' decreases to a small value at $H_0 = H_{c3}$ and to zero at $H_0 = H_{c3}+h_0$. At the maximum value of χ_1'' the corresponding value of $\chi_1' = -0.47/4\pi$. Both χ_1' and χ_1'' have horizontal tangents as a function of H_0 at $H_0 = H_{c3}+h_0$. When for a given material the ac amplitude h_0 is changed, the maximum occurs at a different static field H_0. Hence by changing the amplitude h_0, one can measure $h_c = 0.385 h_0 = 4\pi M_1$ (Ref. 4, Fig. 1) as a function of H_0 and thus obtain direct information on the critical state of the surface sheath from the measured ac losses.

In the above discussion we have assumed that the normal-state susceptibilities are zero. This is, however, not the case in a real metal whose susceptibilities are (H_0 parallel to the axis of the cylinder)[7]

$$\chi_n' + i\chi_n'' = -\frac{1}{4\pi}\left[1-\frac{2}{Rk}\frac{J_1(Rk)}{J_0(Rk)}\right], \tag{10}$$

where $k = (2i)^{1/2}/\delta$, δ is the rf skin depth, and the J's are Bessel functions. χ_n'' reaches a maximum for $R/\delta = 1.8$ whose value $\chi_{n\,\max}'' = 0.291/\pi^2 < \chi_{1\,\max}'' = 0.334/\pi^2$. Thus when the ratio of the radius R to the skin depth δ is most unfavorable the maximum absorption in the superconducting sheath is still about 15% larger than that of the normal state. When $R < \delta$, the normal core of the cylinder enclosed by the superconducting surface sheath will partially contribute to the absorption when $H_0 < H_{c3}+h_0$, thus enhancing the measured peak of χ_1'' with respect to χ_n''.

Let us make some numerical estimates for the sample used in Ref. 1, Fig. 1 (Pb-0.1% Bi at 4.2°K). With $H_c = 540$ G, $H_{c3} = 700$ G, the κ value is 0.54 from which it follows that $\lambda = 5.0\times10^{-6}$ cm. With $f = \omega/2\pi = 18$ cps, $R = 7.62\times10^{-2}$ cm, and $\chi_n'' \approx (\frac{1}{4})\chi_{1\,\max}''$, one obtains[7] two solutions for δ, namely $\delta_1 = 1.01\times10^{-1}$ cm and $\delta_2 = 1.16\times10^{-2}$ cm, and two corresponding solutions for the resistivity of Pb-0.1% Bi, namely $\rho_1 = 1.0\times10^{-2}$ $\mu\Omega$ cm and $\rho_2 \approx 0.5\times10^{-4}$ $\mu\Omega$ cm. These solutions correspond to χ_n' of about $-0.02/4\pi$ and $-0.85/4\pi$, respectively. The latter solution is physically unrealistic as far as both the resistivity ρ_2 and the corresponding value of χ_n' are concerned. Both ρ_1 and the corresponding value of χ_n' are reasonable for the above sample. Therefore, from the ratio of the normal-state absorption to the maximum absorption in the superconducting sheath state, one is able to estimate the resistivity of the superconducting metal in the normal state.

The physical mechanism that causes losses in the superconductor is due to the critical state of the surface sheath which is a well-defined function of the magnetic field H_0 for a given material. When a critical current is flowing in the superconducting surface sheath and H_0 is increased, the critical current must decrease in order to conserve in the time average the condition[4] $G_{SH}-G_{NH}=0$, where the G's are the magnetic Gibbs functions in the superconducting and normal state, respectively. The experimental verification is given in Ref. 3 where it was shown that the calculations of Ref. 4 are satisfied without any adjustable parameters.

As the superconducting surface sheath encloses the mixed state[3,8] it is likely that the above arguments hold also qualitatively for applied magnetic fields between H_{c1} and H_{c2}. It is likely that the critical currents in the superconducting sheath make a contribution to the measured low-frequency losses between H_{c1} and H_{c2} in addition to those due to flux pinning (which are not understood at present).

When the external magnetic field is swept at a constant rate dH_0/dt and no ac magnetic field is superimposed on the swept field, one measures a voltage which is proportional to dM/dH_0. In other words, one measures the slope of the static magnetization curve directly.[3] When a time-varying field $h_0 \sin\omega t$ is superimposed on the swept field[9] the interpretation of the experimental results is more complex, though the physics of the superconducting surface sheath is the same as has been discussed above.

I would like to thank L. J. Barnes for interesting discussions.

Note added in proof.—Recent experiments by P. O. J. Van Engelen, G. J. C. Bots, and B. S. Blaisse [Phys. Letters 19, 465 (1965)] and also by E. Maxwell and W. P. Robbins (to be published) came to the author's attention. These experiments can be readily interpreted with the above theory.

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[5]When H_0 is kept constant and h_0 is slowly reduced from h_c to zero, the $4\pi M$ value of the sample will approximately vanish. The total current in the sheath is then zero and the sample is "demagnetized."

[6]For simplicity we have linearized the $\pm 4\pi M$ curves and the $h_i(t)$ curve. This is a good approximation for samples of the size used in Ref. 1.

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NEAR-FORWARD RAMAN SCATTERING IN ZINC OXIDE

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In polar crystals, electromagnetic waves and optical phonons interact strongly when their energies and wave vectors are nearly equal. This interaction removes the intersection of the uncoupled dispersion curves,[1] and results in an upper and lower branch which have mixed electromagnetic and mechanical nature. The crystal excitations in this mixed region have been called polaritons by Henry and Hopfield[2] who were the first to observe Raman scattering from them. Henry and Hopfield observed a 20% shift in the polariton frequency in GaP as the scattering angle was varied from the forward to near-forward direction.

Because ZnO is a uniaxial crystal, we have been able to obtain nearly a three-fold shift in the polariton frequency as a function of angle in the near-forward direction. The advantage of a uniaxial crystal[3] is that in a positive crystal such as ZnO the frequency of the polariton that conserves energy and wave vector can be made small if one makes the incident light an ordinary ray and the Stokes Raman light an extraordinary ray with the maximum index. The frequency of the allowed polariton is increased as the angle from the forward direction is increased, so that a frequency shift from 160 to 407 cm^{-1} has been obtained.

For comparison, the case in which the input is an extraordinary ray with the maximum index and the Raman light is an ordinary ray has also been studied. Here, the wave vector and therefore the frequency of allowed polariton is large in the forward direction, so that a shift of only around 20% in the polariton frequency was obtained.

Our experimental setup consists of the ordinary (extraordinary) incident light (4880 Å from the ionized argon laser) propagating along the x axis while the extraordinary (ordinary)