= $\overline{m}_{,;}$ so it is possible for the spontaneous moment to change direction without altering the basic antiferromagnetic ordering. Of the two structures, the one that yields the minimum Φ actually exists. Substitution of Eqs. (3) into (2) yields

$$\Phi_{\rm II_{+}} = \Phi_{\rm II_{-}} = \frac{A}{2} l^2 - \frac{\beta^2}{2B} l^2 + \frac{C}{4} l^4, \qquad (4)$$

or the states with $\pm\beta$ are degenerate. If we write this result in the form of Moriya's² notation, we must express the interaction between two ionic spins on a microscopic basis as

$$\pm \mathbf{\vec{d}} \cdot (\mathbf{\vec{s}} \times \mathbf{\vec{s}}_2), \tag{5}$$

which provides for the possibility of both states allowed by Eq. (4) since \bar{d} is allowed to point in either direction along the [111] axis. Experiments indicate that most of the data can be described in the molecular field approximation by including the term given in relation (1). Therefore we conclude that there must be an interaction between the microscopic \bar{d} vectors which tends to align them spontaneously in much the same manner as the spins in a true ferromagnet are aligned. The microscopic origin of the interaction, however, would have to be of a different nature. This leads to some interesting conclusions.

(A) It is possible for a ferromagnetic domain structure to exist in the absence of an antiferromagnetic domain structure. In this case the macroscopic \vec{D} vector would change direction from one domain to another leaving the antiferromagnetic structure unchanged. In the domains the energy density would be the same, excluding interaction with external fields, as has been shown. However the energy density in the domain walls would increase, as is the case for any domain structure. This follows from the decrease in the magnetic symmetry within the domain wall. It is possible that this effect has already been observed.³

(B) The macroscopic \vec{D} vector exists only because of a spontaneous alignment of the microscopic \vec{d} vectors against the randomizing effect of kT; therefore \vec{D} in expression (1) should be replaced by $\langle \vec{D} \rangle = \vec{D} f$, where \vec{D} is the value of $\langle \vec{D} \rangle$ at $T = 0^{\circ}$ and f is the appropriate statistical distribution function. This result may supply the reason for the lack of saturation of the ferromagnetic moment in fields below several thousand Oe while the in-plane anisotropy fields are not larger than ~100 Oe.

(C) Preliminary computer calculations show that by using a statistical value for $\langle \vec{D} \rangle$, better agreement between experiment and theory is obtained for the field-induced transition when the field is applied in the basal plane.

Further studies of these effects are in progress and will be reported in due course.

SURFACE IMPEDANCE OF NIOBIUM NEAR H_{c2} *

W. D. Hibler, III, and B. W. Maxfield

Laboratory of Atomic and Solid State Physics, Cornell University, Ithaca, New York 14850 (Received 28 June 1968)

The magnetic field and angular dependence of the surface impedance of superconducting niobium near H_{c2} is measured and compared with predictions based on the theory of Caroli and Maki.

Recently, Caroli and Maki¹ (CM) have calculated the electromagnetic response for a pure type-II superconductor in the limit of small order parameter Δ . The calculation is performed using linear response theory generalized to include fluctuations in Δ . CM assume that in a pure type-II superconductor near H_{c2} the effect of a

magnetic field on the order parameter is similar to that of a transport current.² This enables them to calculate the retarded commutators in the electromagnetic response function.

Using an approach similar to CM, Maki had previously calculated the ultrasonic attenuation in a pure type-II superconductor.² This approach

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appeared to explain the systematic change in the field dependence of the ultrasonic attenuation as a function of purity that was observed by Gottlieb, Jones, and Garbuny.³ Kagiwada <u>et al.</u>⁴ have determined the temperature dependence of the ultrasonic attenuation in niobium. They obtain a good fit of their data to the Maki theory using a value of the density of states at the Fermi surface, $N(0) = 1.5 \times 10^{34}$ states/erg cm³, which is considerably smaller than the value $N(0) = 5.6 \times 10^{34}$ states/erg cm³ estimated from specific-heat measurements. No measurements of anisotropy in the ultrasonic attenuation have been reported to date.

To provide a further check on Maki's approach to calculating the transport properties of a pure type-II superconductor near H_{c2} , (i.e., the CM theory) we have measured the surface impedance Z in the mixed superconducting state of pure niobium.⁵ The magnetic field dependence of Z is measured in a number of specimens having resistance ratios between 6500 and 11000. Hence, all the specimens studied here satisfy the condition for the pure limit, $l \gg \xi_0$, where l is the electronic mean free path and ξ_0 is the coherence length. These resistance ratios are considerably larger than those used by the previous workers.^{3,4} In fact, one expects the purity dependence of the order parameter to be only a few percent over the range of purities studied here. Therefore, in these specimens, the magnetic field dependence of Z is not expected to vary with the electronic mean free path.

We compare the measured surface impedance with calculations based on the electromagnetic response functions of CM. As in the case of the ultrasonic attenuation measurements,^{3,4} agreement in functional form for the magnetic field dependence is obtained. We have also measured the anistropy of the surface impedance. In particular, we keep the magnetic field H_0 perpendicular to the induced currents J and vary the angle between H_0 and q, the direction of decay of the screening currents. For all measurements discussed here, q is parallel to \hat{n} , the specimen normal (see insert of Fig. 2). The anisotropy results reported in this Letter do not agree with numerical calculations that we have done, based on the CM response functions.⁵

Both real and imaginary parts of Z were measured using a standard twin-tee rf bridge by setting the bridge out of balance in either a resistive or reactive direction. Relatively large off-balance signals were used to avoid mixing

real and imaginary components in the detected output. The off-balance signals were not so large that the bridge linearity was affected; that is, the detected output was proportional to the corresponding impedance change (resistance or reactance). These precautions were necessary since the real and imaginary parts of Z change quite rapidly near H_{c2} . A marginal oscillator was also used to measure Z and the results agreed well with the bridge method. All results reported in this Letter were obtained using the bridge method.

To determine the magnetic field dependence of Z, the detected output of the bridge was plotted as a function of the magnetic field on an X-Y recorder. The angle θ between H_0 and the sample normal could be varied from 0° to 360°. Data were recorded by setting the desired angle and sweeping the field slowly (about 300 G/min) from zero to well above H_{C2} . No hysteresis was observed for $H_0 > \frac{1}{2}H_{C2}$. We observed a detected signal that depended upon the sweep rate for speeds faster than about 300 G/min. These ratedependent effects were not related to instrument response times.

The anisotropy in Z was determined from swept-field measurements at a number of different values of θ . This proved superior to plotting the detected output versus θ at a fixed H_0 because the single-crystal specimens exhibited a θ -dependent H_{C2} ⁶ and a θ -dependent normalstate surface impedance (due to the anisotropy of $V_{\mathbf{F}}$).

Using the CM response functions appropriate to the experimental geometry shown in the insert of Fig. 2 (that is $H_0 \perp J$), we find that the extreme anomalous surface impedance for specular scattering is given by (following CM we use units where $\hbar = k_{\rm B} = c = 1$)

$$\frac{Z_s}{Z_n} = 2 \left[\frac{\exp(i\pi)}{D(\theta, \rho)} \right]^{1/3},$$

where

i

$$D(\theta, \rho) = 1 - \left(\frac{\Delta}{2T}\right) \left[1 - \ln \frac{8\Delta}{|\omega|}\right] G(\theta, \rho),$$

$$\rho=\epsilon/2\pi T,\ \epsilon=V_{\rm F} \left[{\textstyle \frac{1}{2}} e H_{c2} \right]^{1/2}, \label{eq:rho}$$

and

VOLUME 21, NUMBER 11

$$G(\theta,\rho) = 4V_{\mathbf{F}} \int_{0}^{1} dZ [1-Z^{2}]^{1/2} \int_{-\infty}^{\infty} \frac{e^{-x^{2}/(1-Z^{2}\sin^{2}\theta)}}{[1-Z^{2}\sin^{2}\theta]^{1/2}} \cosh^{-2}(\pi\rho x) \frac{dx}{\sqrt{\pi}}.$$

For $\theta = 0$ and $\frac{1}{2}\pi$, $G(\theta, \rho)$ reduces to the functions $f_{\perp}^{(1)}(\rho)$ and $f_{\perp}^{(2)}(\rho)$, respectively, defined by CM. We modify the Maki expression for the order parameter² to include an adjustable parameter α . That is, we use

$$\Delta^{2} = \frac{\alpha}{2N(0)\pi} \frac{H_{c2}^{-H_{0}}}{\beta [2K_{2}^{-2}(T) - 1]} \left[H_{c2}^{-\frac{1}{2}}T \frac{dH_{c2}}{dT} \right], \quad \beta = 1.16,$$

where $K_2(T)$ is the second Ginsberg-Landau parameter. With $\alpha = 1$ this is the order parameter used by Kagiwada <u>et al.</u>⁴ We determined $K_2(T)$ and $H_{C2} - \frac{1}{2}T(dH_{C2}/dT)$ from the results of McConville and Serin⁷ and used $N(0) = 5.6 \times 10^{34}$ states/cm³ erg, the result obtained from specific heat measurements.^{7,8} Using these values and an estimate for the Fermi velocity, $V_{\mathbf{F}} = 9 \times 10^7$ cm/sec, a computational program was carried out to determine the value of α which gave the best fit to the $\theta = 0^\circ$ experimental data near H_{C2} . $G(\theta, \rho)$ was evaluated on an IBM 360 computer. As illustrated by the dashed curve in Fig. 1(a), we obtain a reasonable fit with the experimental results at 4.2°K using $\alpha = 0.92$. Expressed more directly in terms of the normalized surface impedance, this fit corresponds to

$$(\Delta/2T)G(0^{\circ},\rho) = B[(H_{c2}-H_{0})/H_{c2}]^{1/2} = 0.18[(H_{c2}-H_{0})/H_{c2}]^{1/2}$$

Calculations carried out using the expression for Δ^2 in Ref. 1 yield a value of *B* that is too large by a factor of more than 100.

Swept-field data for $\theta = 0^{\circ}$ was also taken at 1.95°K. For fixed H_0/H_{c2} , the theory predicts a decrease in the parameter B from its value at 4.2° K by a factor of 0.78. The best fit of the theory to the experimental data at 1.95°K corresponds to a decrease in B by a factor of 0.72. Curves 1 and 2 in Fig. 1(b) illustrate the fit to the experimental results at 4.2 and 1.95°K obtained by using the theoretical factor 0.78. Note that data over a much larger field range are shown. As is evident from curve 3 in Fig. 1(b), a less pure specimen requires a larger value of B to fit the experimental data to the theory. This is in direct disagreement with the theory. The only theoretical parameter that has any purity dependence is K_2 , and this variation is much too small to explain the observed effect. Note that we measure H_{C2} so that it is effectively an experimental parameter. This too exhibited a very small (< 2%) purity dependence over the specimen range studied.

For a fixed H_0 about 100 G below H_{c2} , the measured surface resistance as a function of θ is shown by the solid curve in Fig. 2. The angular dependence is qualitatively the same for a wide range of fields below H_{c2} . The dashed curve represents the CM theory and is determined by numerical calculations of $G(\theta, \rho)$ at values of θ represented by the circles.⁵ As mentioned previously, the analysis of angular data is complicated by an observed angular dependence in H_{c2} .⁶ For Nb-5 we observe a 2% change in H_{c2} which of course also causes a change in dM/dH at H_{c2} . These changes were neglected in calculating the dashed curve. However, even taking a 5% change in H_{c2} at $\theta = 45^{\circ}$ and carrying out the calculations including these effects, the predicted surface resistance still decreases as a function of θ (but the decrease is slightly small-er).

The theory implicitly assumes a spherical Fermi surface. Since the angular dependence predicted by the theory is rather small, it is conceivable that an anisotropic normal-state conductivity could account for the discrepancy between the observed and predicted angular dependence. However, we point out that most of this angular variation is already taken into account because the mixed-state surface resistance is normalized to the normal-state value. In addition, one would not expect such a monotonic angular variation of the normal-state conductivity over such a large angular range about a nonsymmetry axis.

The observed increase in R_S/R_n would be less in the presence of a superconducting surface sheath because any effect of the surface sheath, which tends to lower the resistance, will increase as θ increases from zero. We expect the influence of the surface sheath to be small over the angular range shown. However, if these effects are present, it means that the actual bulk properties exhibit an angular dependence greater than that observed in the experiments.



FIG. 1. Field dependence of the normalized mixedstate surface resistance of niobium near $H_{c2^{\circ}}$. In (a) the circles are experimental data points while the dashed curve was determined numerically using α = 0.92 or B = 0.18. In (b) we show the effect of temperature and purity on the surface resistance over a larger field region than shown in (a). Curves 1 and 2, calculated using $\alpha = 1.38$, reflect the temperature dependence predicted by CM. The data points, circles and squares, are in fair agreement. Curve 3, corresponding to $\alpha = 2.74$ or B = 0.31, illustrates an effect of purity on the surface resistance not predicted by CM.

In summary, the observed field dependence of the surface resistance near H_{C2} is found to have the same functional form as predicted by the CM theory. Similar agreement was observed in previous^{3,4} ultrasonic-attenuation measurements. The order parameter appears to have a meanfree-path dependence not contained in the theory. This fact was not evident from the ultrasonic-attenuation measurements. The larger values of α required to fit our data for the less pure specimens are consistent with the observation of Kagiwada et al.⁴ that a rather small value of N(0) was



FIG. 2. Relative change in the normalized surface resistance as the angle between H_0 and q is varied. All directions are defined by the insert. The solid curve is drawn through the experimental data represented by the solid circles. The dashed curve is drawn through the circles which are computed using the CM theory. Within experimental error, the results are symmetrical about $\theta = 0^{\circ}$.

necessary to obtain a good fit to their data. Over the rather limited temperature range that we studied, the CM theory gives a reasonable prediction of the observed temperature dependence. The CM theory does not explain the observed anisotropy in the surface impedance.

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