NUCLEATION OF SUPERCONDUCTIVITY IN TANTALUM IN A DECREASING MAGNETIC FIELD

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Measurements of the surface nucleation field H_{c3} and the bulk supercooling field H_{sc}, made on pure tantalum, show that these fields are identical only when $\kappa < 0.39$, and that measurable supercooling occurs at all temperatures between T_c and 1.4°K. The measurements verify qualitatively the recent theoretical work of Park. Nearly ideal supercooling of the bulk cylindrical samples is achieved by using small auxiliary-field coils to field coils to eliminate nucleation at the sample ends. A new value for the Ginzburg-Landau parameter of the pure metal $\kappa_0 = 0.340 \pm 0.01$ is derived from the data.

There has recently been renewed interest in superheating and supercooling of superconductors.[1] This is mainly due to recent experiments on superconducting colloids,[2] which avoid most of the difficulties of earlier work[3] on bulk materials, and the success of the Ginzburg-Landau (GL) theory[4] in relating the phenomena to the characteristic parameter $\kappa = \lambda/\xi$. λ is the weak-field penetration depth and ξ is the temperature-dependent coherence length. The nucleation fields for superconductivity are found by solving the GL equations. The minimum field at which spontaneous nucleation occurs in the bulk of the material far away from its surface can be shown to be $H_{c2} = \sqrt{2}\kappa H_c$. This field was at first thought to be the limiting field for supercooling of the normal phase which we denote by H_{sc}. However, Saint-James and de Gennes[5] showed that in a decreasing field nucleation of superconductivity takes place first at the surface when the field reaches a value $H_{c3} = 2.4\kappa H_c$, the field being parallel to the surface. They suggested that when $H_{c3} < H_c$ ($\kappa < 0.417$), H_{c3} should represent the supercooling field. It is assumed, if only implicitly, that the superconducting surface layer is unstable below H_c. According to this hypothesis the values of κ derived from the supercooling experiments of Faber[3] must be lowered from $H_{sc}/\sqrt{2}H_c$ to $H_{sc}/2.4H_c$. However, we have observed experimentally[6] on pure lead and tantalum that supercooling can take place even when $H_{c3} > H_c$. Extending the theoretical work of Feder,[7] one of us[8] explained these results by studying the dependence of the surface order parameter on field and κ from numerical solutions to the GL equations. He was able to show that the normal phase is unstable below H_{c3} when $\kappa < 0.407$, i.e., when $H_{c3} < 0.98H_c$, but that for $\kappa > 0.407$ the supercooling field H_{sc} is less than H_{c3}. As κ increases the calculations indicate that H_{sc} tends to H_c until at $\kappa \simeq 0.6$ there is no appreciable supercooling even though the surface energy remains positive[9] for $\kappa < 0.707$. The value of $\kappa = 0.407$ below which $H_{sc} = H_{c3}$, obtained in the above theory, has been confirmed recently by the calculations of Christiansen and Smith.[10]

As a test of this theory, we have measured the surface nucleation field $H_{c3} > H_c$ and the supercooling field $H_{sc} < H_c$ of a single-crystal tantalum rod over a wide range of temperature. Magnetization measurements were performed as described in Ref. 6 with the exception that to achieve nearly ideal supercooling of a cylindrical sample it was necessary to use small auxiliary coils to avoid nucleation at the sample ends as shown in the inset of Fig. 1. The specimen reported on here was an electron zone-refined monocrystalline tantalum rod 3.9 cm long and 1.9 mm in diameter. The rod axis was determined to be a few degrees from the [111] direction. Pure tantalum is ideally suited for these measurements since, as a result of the temperature dependence of κ, H_{c3} is greater than H_c at low temperatures (T 3.5°κ), less than H_c at T_c (4.482°κ), and equal to H_c at a temperature close enough to T_c for one to hope that the GL theory might apply. If the theory is correct we expect the extrapolation of the curve of $H_{c3}(T)$ to join smoothly onto the $H_{sc}(T)$ curve at $\kappa = 0.407$.

Experimental data on the surface nucleation field $H_{c3} > H_c$ are represented by the solid circles in Fig. 1 corresponding to values of the ratio H_{c3}/H_c, where H_c is measured independently at each temperature. Similarly, the supercooling field is displayed by plotting the ratios H_{sc}/H_c at each temperature before energizing the auxiliary-field coils (triangles in Fig. 1) and after energizing the field coils (open circles). As seen in Fig. 1, a large increase in the degree of supercooling is realized by the use of the auxiliary-field coils. Presumably this is due to the elimination of nucleation of the superconducting phase at the sample ends since the current in these coils was such that the field at the ends was higher than at the

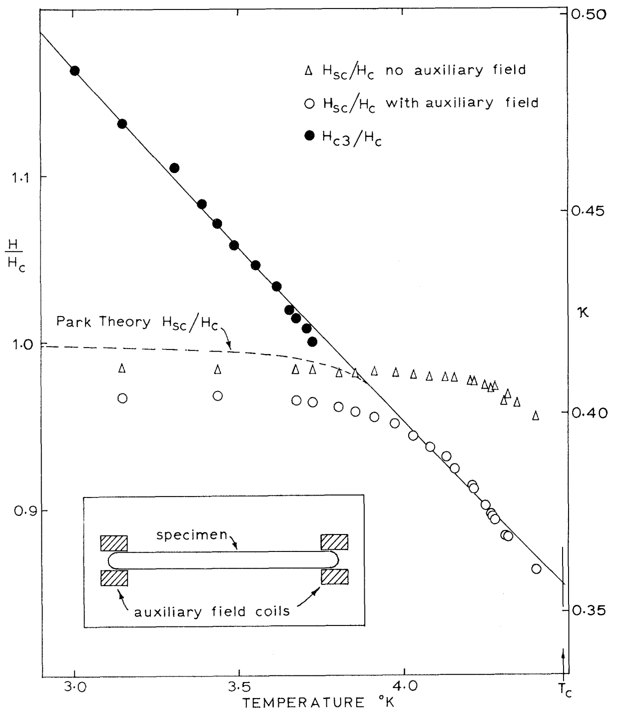

FIG. 1. Effect of auxiliary-field coils placed at the ends of the specimen (see inset) on the observed supercooling field H_{sc}. The specimen is an electropolished single-crystal cylinder of tantalum. The full line is a curve of H_{c3}/H_c obtained from the measurements, as described in the text. The κ scale applies only to this curve.

center of the sample. In taking the data the auxiliary field was gradually increased until no further increase in H_c-H_{sc} was observed. It was then assumed that the transformation to the Meissner state which takes place by the spontaneous growth in thickness of the sheath starts near the center as it would in an ideal specimen. It is presumed that flaws near the ends of the sample reduced the degree of supercooling before the field of the auxiliary coils was applied. The experimental results provide support for the validity of these assumptions, as we point out below.

The effect of the auxiliary field is shown in the sequence of Fig. 2. Note the change in H_{sc}/H_c in Fig. 1, the new values being represented by open circles. The value of H_{sc}/H_c at any given temperature has decreased sharply. Supercooling has increased, particularly near T_c, indicating that in this specimen the regions of the cylinder little affected by the auxiliary coils may be free of nucleation centers. Clearly a reasonable interpolation can be made between the H_{c3}/H_c and the H_{sc}/H_c points which was not possible for the data obtained without the field coils.

The solid curve in Fig. 1 is obtained from two analytic functions which give good fits to the H_{c3}[11] and H_c[12] data where the numerical constants have been obtained by the method of least squares. Near T_c the measured H_{sc}/H_c points

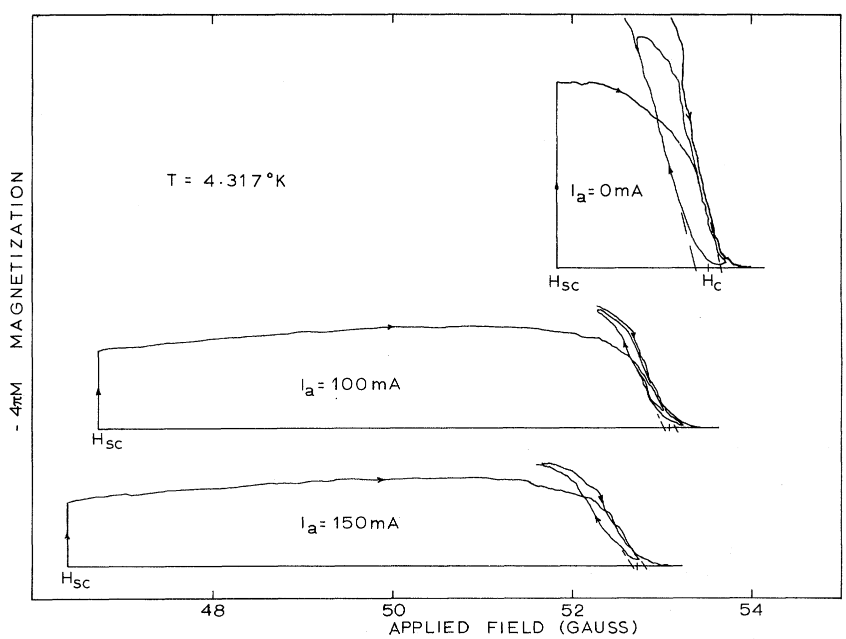

FIG. 2. Effect of the current in the auxiliary-field coils (I_a) on the magnetization curve. Supercooling has been avoided in parts of the curves by reversing the field sweep at the foot of the ascending field magnetization curve in order to obtain the intermediate state hysteresis. This information is necessary in order to estimate H_c by the method described in Ref. 6. The magnetic field scale has been much more expanded than is customary and the magnetization scale contracted, in order to make the effect of I_a clear.

fall very close to the H_{c3}/H_c curve; that is, $H_{c3} = H_{sc}$ when $\kappa \geqslant 0.39$. Although this is 5% lower than Park's theoretical value 0.407, it is in agreement with the corresponding value of Smith and Cardona[13] obtained on Pb and SnIn-alloy spheres suggesting that a numerical discrepancy may exist in the calculation due to simplifying assumptions in the theoretical treatment.

The value of $\kappa(T_c)$ has been determined from the fitted (solid) curve of Fig. 1. The κ scale on the right ordinate applies only to this curve. By κ we mean $\kappa \equiv 0.417 H_{c3}/H_c$. The upper and lower limits are obtained by using graphical extrapolation of curves down through the data points, giving $\kappa(T_c) = 0.36 \pm 0.01$. Using our experimental values of $\kappa(T_c)$ and the residual resistivity ρ_0, we obtain $\kappa_0 = 0.34 \pm 0.01$ from the Gor'kov-Goodman equation.[15] This value is considerably lower than the value $\kappa_0 = 0.42 \pm 0.02$ estimated by Buchanan, Chang, and Serin[16] from upper-critical-field measurements on impure (type 2) tantalum. This difference is somewhat larger than the variation of κ_0 expected from Fermi surface anisotropy.[17] Using our value of κ_0 and the original GL definition[4] $\kappa = 2\sqrt{2}\pi H_c(T)\lambda^2(T)/\varphi_0$, where $\varphi_0 = hc/2e$, along with the BCS expression[18] $\lambda(T) = \lambda_L(0)/[2(1-t)]^{1/2}$ we obtain the London penetration depth of pure tantalum $\lambda_L(0) = 330 \pm 10$ Å. From Gor'kov's result[19] $\kappa = 0.96\lambda_L(0)/\xi_0$ we obtain the coherence length in the pure limit, $\xi_0 = 925 + 40$ Å. We have used $dH_c/dT = 330 \pm 10$ G/deg obtained from our results, and the transition temperature for the pure material $T_{c0} = 4.482°$.[20]

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[1]See J. P. Burger and D. Saint-James in "Superconductivity," edited by R. D. Parks (Marcel Dekker, Inc., New York, to be published).
[2]J. Feder, S. R. Kiser, and F. Rothwarf, Phys. Rev. Letters 17, 87 (1966).
[3]T. E. Faber, Proc. Roy. Soc. (London), Ser. A 231, 353 (1955).
[4]V. L. Ginzburg and L. D. Landau, Zh. Eksperim. i Teor. Fiz. 20, 1064 (1950).
[5]D. Saint-James and P. G. de Gennes, Phys. Letters 7, 306 (1963).
[6]J. P. McEvoy, D. P. Jones, and J. G. Park, Solid State Commun. 5, 641 (1967).
[7]J. Feder, Solid State Commun. 5, 299 (1967).
[8]J. G. Park, Solid State Commun. 5, 645 (1967).
[9]P. G. de Gennes, Superconductivity of Metals and Alloys (W. A. Benjamin, Inc., New York, 1966), p. 207.
[10]P. V. Christiansen and H. Smith, Phys. Rev. 171, 445 (1968).
[11]G. Fischer, Phys. Rev. Letters 20, 268 (1968).
[12]T. P. Sheahan, Phys. Rev. 149, 368 (1966).
[13]F. W. Smith and M. Cardona, Phys. Letters 25A, 671 (1967).
[14]J. P. McEvoy, D. P. Jones, and J. G. Park, to be published.
[15]B. B. Goodman, IBM J. Res. Develop. 6, 63 (1962).
[16]J. Buchanan, G. K. Chang, and B. Serin, J. Phys. Chem. Solids 26, 1183 (1965).
[17]I. Williams and A. M. Court, to be published.
[18]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).
[19]L. P. Gor'kov, Zh. Eksperim. i Teor. Fiz. 36, 1918 (1959) [translation: Soviet Phys.—JETP. 9, 1364 (1959)].
[20]J. Budnick, Phys. Rev. 119, 11578 (1960).

MAGNETIZATION-DENSITY-WAVE STATE IN METALS*

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The thermodynamic potential of a metal with an arbitrary Fermi surface in the presence of a weakly nonuniform magnetic field is evaluated. From it an integral equation for the spatially varying magnetization is obtained, and its solutions are studied.

The possibility of a magnetization-density-wave (MDW) ground state of diamagnetic metals in the presence of a large uniform magnetic field was first suggested[1] by the occurrence of a divergence of the wave-number–dependent diamagnetic permeability $\mu(q)$ at finite values of q. It was soon realized,[2] however, that for a simple spherical Fermi surface, this divergence always occurred first at zero wave number. This situation, in which $\mu(0)$ diverges, is thermodynamically absolutely unstable, and leads to a stratification into diamagnetic domains[3] instead of a MDW state. Recently, Azbel'[4] has pointed out that for a material with more than one extremal cross section of the Fermi surface normal to the magnetic field, the divergence of $\mu(q)$ can oc-