

Critical-Field Curves for Gapless Superconductors*

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Superconducting critical-field curves have been measured for pure Th and Th-Gd alloys at magnetic-impurity concentrations up to 60% of the critical concentration required to completely quench superconductivity. These samples all show a Meissner effect and type-I magnetic transitions which are reversible to at least 1% of H_c . The Abrikosov-Gor'kov theory has been used to make detailed calculations of the critical-field curves over the entire temperature range, and of lifetime broadening up to $\Gamma/\Delta(0, \Gamma) = 1$. Theory and experiment agree to an accuracy of better than 0.5%.

INTRODUCTION

ELECTRON-TUNNELING measurements on superconducting thin films have provided a strong confirmation of the Abrikosov-Gor'kov¹ (AG) prediction of gapless superconductivity. Reif and Woolf² have studied the case in which gaplessness is induced by magnetic-impurity scattering and more recently Hauser³ has studied the case in which gaplessness arises from the proximity effect of magnetic overlays. Both of these experiments qualitatively support the theory but the measurements tend to show a higher density of states than the predicted value. Probably the best quantitative support for the theory comes from the experiments of Millstein and Tinkham,⁴ in which gaplessness is induced by nonmagnetic-impurity scattering in an external magnetic field. Here the experiments agree with theory to an accuracy of a few percent if correction is made for mean-free-path effects. In a somewhat different but related experiment, Mochel and Parks⁵ have studied the transport properties of type-II superconductors in the surface sheath regime and here again the agreement with theory is good. Each of these measurements depends on surface phenomena and for these situations there is a sound experimental foundation for the AG theory.

In bulk materials, however, the experiments on gapless superconducting have been less convincing, primarily because there were serious sample-preparation problems. Specific-heat measurements⁶ for the lanthanum-rare-earth system qualitatively showed the effect but the transitions were rather broad and the interpretation was somewhat ambiguous. Magnetization measurements on these same samples permitted a determination of the upper critical field H_{c2} ,⁶ but the phase transitions were so irreversible that bulk critical-

field data were not reliable. More recent critical-field results by Sugawara and Eguchi⁷ are a great improvement but reversibility is still a problem.

In many ways thorium is more satisfactory than lanthanum as the host superconductor for these measurements. The magnetic rare-earth elements are readily soluble in concentrations up to 60%⁸ so there are no solid solution problems near 1% impurity. In addition, there are no difficulties with mixed phases or multiple crystal structures as in La. Thorium has a relatively low transition temperature⁹ but this is a desirable feature for alloy work because low T_c implies low critical-impurity concentrations¹ and hence the material will remain a type-I superconductor over much of the range of interest. Another advantage of low concentrations of magnetic impurity is that the magnetic ordering temperature will be very low and there will be no complications from this effect. A preliminary measurement of magnetic susceptibility shows that the magnetic ordering temperature in the Th-Gd system increases approximately 0.4°K/at.% Gd, so the magnetic coupling is rather weak. The magnetic ordering temperature is well below the temperatures used here.¹⁰

Detailed critical-field data are presented here for pure Th and Th-Gd alloys at concentrations up to 0.2 at.% Gd and the results are analyzed in terms of deviations from the AG theory. A whole family of critical-field curves have been calculated within the theory to show the detailed dependence of the critical field on the lifetime-broadening parameter Γ . We will discuss the calculations first and then proceed to the experiments.

CALCULATION OF $H_c(T, \Gamma)$

Skalski *et al.*¹¹ (SBW) have extended the AG theory to calculate several of the thermodynamic functions

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¹ A. A. Abrikosov and L. P. Gor'kov, *Zh. Eksperim. i Teor. Fiz.* **39**, 178 (1960) [English transl.: *Soviet Phys.—JETP* **12**, 1243 (1961)].

² M. A. Woolf and F. Reif, *Phys. Rev.* **137**, A557 (1965).

³ J. J. Hauser, *Phys. Rev.* **164**, 558 (1967).

⁴ J. Millstein and M. Tinkham, **158**, 325 (1967).

⁵ J. M. Mochel and R. D. Parks, *Phys. Rev. Letters* **16**, 1156 (1966).

⁶ D. K. Finnemore, D. L. Johnson, J. E. Ostenson, F. H. Spedding, and B. J. Beaudry, *Phys. Rev.* **137**, A550 (1965).

⁷ T. Sugawara and H. Eguchi, *J. Phys. Soc. Japan* **23**, 965 (1967).

⁸ M. Norman, I. R. Harris, and G. V. Raynor, *J. Less-Common Metals* **11**, 395 (1966).

⁹ J. E. Gordon, H. Montgomery, R. J. Noer, G. R. Pickett, and R. Torbon, *Phys. Rev.* **152**, 432 (1966).

¹⁰ D. T. Peterson, D. F. Page, R. B. Rump, and D. K. Finnemore, *Phys. Rev.* **153**, 701 (1967).

¹¹ S. Skalski, O. Betbeder-Matibet, and P. R. Weiss, *Phys. Rev.* **136**, A1500 (1964).

and their work provides the foundation for these calculations. The free energy is given by

$$F_S - F_N = -N_0 \{I_1 + I_2 + \Delta^2\} + \Delta^2/V$$

and the critical field is

$$H_c = [H_{0p} \sqrt{2} / \Delta_p(0)] \{I_1 + I_2 + \Delta^2(T, \Gamma) (1 - 1/N_0 V)\}^{1/2},$$

where

$$I_1 = \int_0^{\omega_0'} d\omega \left(\frac{N(\omega)}{N_0} - 1 \right) 2\omega \tanh \frac{1}{2} \beta \omega,$$

$$I_2 = \frac{4}{\beta} \int_0^{\omega_0'} d\omega \left(\frac{N(\omega)}{N_0} - 1 \right) \left[\ln(1 + e^{-\beta \omega}) + \frac{\beta \omega}{e^{\beta \omega} + 1} \right],$$

$$\omega_0' = (\omega_D^2 + \Delta^2(T, \Gamma))^{1/2},$$

H_{0p} is the pure metal critical field at $T=0$, N_0 is the normal-state density of states at the Fermi surface, $\Delta(T, \Gamma)$ is the order parameter, and $\beta = (k_B T)^{-1}$.

To evaluate these integrals it is first necessary to determine Γ from the ratio of T_c/T_{cp} by the equation

$$\ln T_c/T_{cp} = \psi(\frac{1}{2}) - \psi(\frac{1}{2}[\rho + 1]),$$

where

$$\rho = T_{cp} \Gamma / T_c \gamma_e \Delta_p(0),$$

$\ln \gamma_e$ is the Euler constant, and ψ is the digamma function. An alternate method is to solve

$$(N_0 V)^{-1} = \int_0^{\omega_0'} d\omega \frac{\omega}{\omega^2 + \Gamma^2} \tanh \frac{1}{2} \beta_c \omega,$$

where

$$\beta_c = (k_B T_c)^{-1},$$

for Γ . The results of these two methods agree to 0.01% of Γ . To solve for the order parameter $\Delta(T, \Gamma)$ we have followed the work of Baratoff,¹² where one defines

$$t = T/T_c, \quad \bar{\Gamma} = \Gamma/\Delta(T, \Gamma),$$

$$\delta = \Delta(T, \Gamma)/\Delta_p(0), \quad \delta = \gamma_e t/x_0,$$

$$x_0 = (2n+1)x_0, \quad x_n = \tilde{x}_n [1 - \bar{\Gamma}/(1 + \tilde{x}_n^2)^{1/2}],$$

and iteratively solves the equation

$$\ln t = 2x_0 \sum_{n=0}^{\infty} [(1 + \tilde{x}_n^2)^{-1/2} - (\tilde{x}_n + \bar{\Gamma})^{-1}] - [\psi(\frac{1}{2}[\bar{\Gamma}/x_0 + 1]) - \psi(\frac{1}{2})]$$

for x_0 . $\Delta(T, \Gamma)$ then follows directly from $\Delta(T, \Gamma) = \Delta_p(0) (\gamma_e t/x_0)$. Usually 10 to 50 iterations are required to obtain $\Delta(T, \Gamma)$ to an accuracy of 0.01%. The remaining parameter in the calculation is the density of states and this is given by

$$N(\omega)/N_0 = (1/\bar{\Gamma}) \text{Im} u,$$

¹² A. Baratoff, Ph.D. thesis, Cornell University Report No. 256 (unpublished).

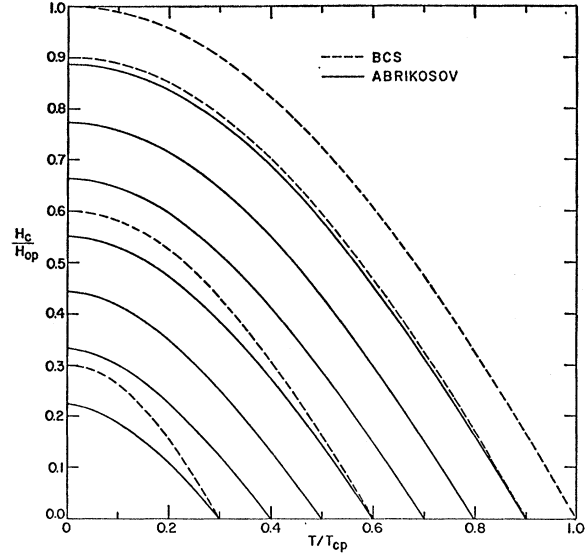


FIG. 1. Comparison of the BCS and the AG predictions for the critical-field curves. BCS would presumably apply for nonmagnetic impurities and AG would apply for magnetic impurities.

where u is the solution of the quartic equation

$$u^4 - 2\bar{\omega}u^3 + (\bar{\omega}^2 + \bar{\Gamma}^2 - 1)u^2 + 2\bar{\omega}u - \omega^2 = 0.$$

With these values for Γ , $\Delta(T, \Gamma)$, and $N(\omega)/N_0$, one can evaluate the integrals for $H_c(T, \Gamma)$.

The resulting critical-field curves, which were calculated for eight values of Γ at intervals of 0.1 of T_c/T_{cp} , are shown in Fig. 1. The curves remain nearly parabolic in shape with a monotonically decreasing ratio of H_0/T_c as T_c/T_{cp} decreases. If the results are cast as deviations from a parabolic law (Fig. 2), the deviations become more negative as Γ increases and, for a value of $\Gamma/\Delta_p(0) = 0.378$, the magnitude of the deviation is approximately -7% . More complete details of the calculation have been discussed by Decker.¹³

EXPERIMENTAL PROCEDURE

Cryostat

A ballistic induction technique was used to obtain magnetization data and hence the critical-field curves for these samples. The apparatus, shown in Fig. 3, is similar to one described earlier, so only the essential features will be reported here. Temperatures from 0.3 to 1.5°K were provided by a conventional He³ refrigerator in which temperature could be controlled to 0.0001°K. In order to establish a temperature scale a germanium resistance thermometer was calibrated

¹³ W. R. Decker, Ph.D. thesis, Iowa State University (unpublished).

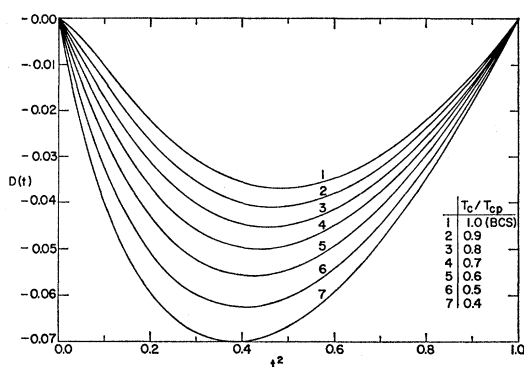


FIG. 2. Deviation functions for the AG critical-field curves. Deviations from a fiducial parabola become more negative as the lifetime broadening increases.

against the vapor pressure¹⁴ of He^4 from 4.2 to 1.3°K and against the susceptibility of cerium magnesium nitrate (CMN) below 1.3°K. The CMN was ground to a particle size of 0.5 mm, mixed with 50 centistokes silicone oil, and pressed into a spherical chamber. Susceptibilities were measured with a 33-Hz mutual inductance bridge which gave temperatures to a precision of $T^2/8000^\circ\text{K}$. No correction was made to the Curie-law behavior in accord with the results of other investigators.^{15,16}

After the germanium resistance calibration was completed, the CMN salt pill was replaced by the apparatus for the superconducting magnetization measurements as shown in Fig. 3. This consisted of three counter-wound pickup coils with 64 000 turns of No. 46 wire which were symmetrically placed around the axis of the cryostat and thermally connected to the He^3 refrigerator with a copper yoke. The germanium resistance thermometer and one end of each sample was affixed into holes in the copper yoke with Apiezon N grease or GE7031 varnish to provide good thermal contact. The free end of each sample extended into the pickup coils so that most of the signal came from unstrained portions of the specimen. A shield at the temperature of the He^3 refrigerator surrounded the entire apparatus.

A sixth-order Garrett¹⁷ solenoid, which was used to provide the main field, was homogeneous to 0.01% over the region of the samples. The nuclear magnetic resonance of protons in glycerine was used to calibrate the solenoid.

Sample Preparation

Sample preparation is crucial in these experiments. The samples must be sufficiently homogeneous to

¹⁴ F. G. Brickwedde, H. van Dijk, M. Durieux, J. R. Clement, and J. K. Logan, *J. Res. Natl. Bur. Std. A* **64**, 1 (1960).

¹⁵ R. P. Hudson and R. S. Kaeser, *Physics* **3**, 95 (1967).

¹⁶ W. R. Abel, A. C. Anderson, W. C. Black, and J. C. Wheatley, *Physics* **1**, 337 (1965).

¹⁷ M. W. Garrett, *J. Appl. Phys.* **22**, 1091 (1951).

show sharp phase transitions with little hysteresis so that the critical field is a well-defined parameter. Even though the samples reported here are very good in this respect, the sample quality still limits the accuracy of the measurement.

Two methods were used to prepare pure Th. An electrotransport¹⁸ process in vacuums of 10^{-9} Torr gives samples with resistivity ratios of approximately 1200.¹⁰ All the pure Th critical-field data presented here were taken with a sample prepared by this method.

Unfortunately, electrotransport cannot be used to prepare the alloys because Gd atoms are swept out of the sample at rates comparable to that of carbon and oxygen. Hence, a second pure Th sample and the alloys were prepared by a conventional arc melting and annealing method. Table I gives a partial chemical analysis of the starting Th. A master alloy of Th 5.0 at. % Gd was prepared by arc melting and the other samples were formed by diluting the master alloy with pure Th. Each sample was arc melted four times to insure homogeneity. The weight changes in arc melting were less than 0.03%, so no significant amount of Gd was lost. The arc-melted button was then swaged into wires 0.060 in. in diameter wrapped in Ta foil and annealed at 800°C for $\frac{1}{2}$ h under vacuum to allow

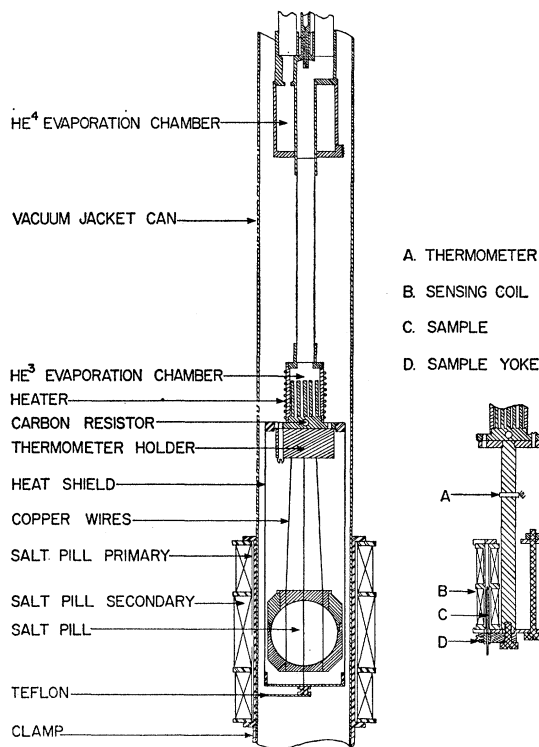


FIG. 3. Cryostat drawing.

¹⁸ D. T. Peterson, F. A. Schmidt, and J. D. Verhoeven, *Trans. AIME* **236**, 1311 (1966).

recrystallization. The superconducting transitions of these samples usually showed considerable hysteresis, so the samples were wrapped in thorium foil, sealed in a Ta container, and annealed again at 1200°C for 7 days. These samples then usually exhibited satisfactory superconducting transitions.

At concentrations greater than 0.20 at.% Gd all the samples showed a great deal of hysteresis and even T_c showed rather erratic behavior. At this time it is not known why the difficulties arise but it may be related to a transformation from type-I to type-II behavior.

RESULTS AND DISCUSSION

Pure Thorium

Two samples of pure thorium have been studied in detail and the results are roughly equivalent, despite a wide difference in resistivity ratio $R_{300\text{ }^\circ\text{K}}/R_{4.2\text{ }^\circ\text{K}}$ from 1200 to 35. For the 1200-resistivity-ratio sample, the shape of the transitions is typical of long cylindrical specimens, as shown in Fig. 4. In the intermediate state the transitions are completely reversible but, once

TABLE I. Analysis of thorium used to prepare samples (ppm).

| | |
|----------------|-----|
| Carbon | 25 |
| Oxygen | 110 |
| Nitrogen | 20 |
| Iron | 5 |
| Other metallic | <20 |

the sample has gone completely normal and the magnetic field reduced, there is supercooling of about 0.68% of H_c . The general character of this supercooling is common in high-purity materials and generally indicates a high-quality sample. If it is assumed that the true nucleation field is given by these hysteresis measurements, then the value of κ ¹⁹ for thorium would be 0.41.

The high-resistivity pure Th sample showed no supercooling on the initial run (after it had been annealed at 800°C for ½ h) but a subsequent anneal at 1200°C for 7 days sharpened the transitions and apparently changed the nucleation properties so that the sample showed supercooling of about 0.5% of H_c . The critical-field data for this sample parallel those for the 1200-resistivity-ratio sample, with about 4G separation in the measurement, so they won't be reported here. The arc melting and annealing apparently introduced enough impurity to depress T_c by 0.026°K.

The critical-field curve for pure thorium, like most superconductors, closely follows a parabolic temperature dependence so the data are displayed as deviations from a fiducial parabola in Fig. 5. At all temperatures

¹⁹ D. St. James and P. G. de Gennes, Phys. Letters 7, 306 (1963).

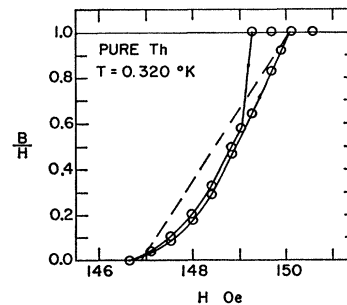


FIG. 4. Magnetic phase transition for the pure Th sample with a 4°K resistivity of 0.0128 $\mu\Omega$ cm.

the critical-field curve of Th lies within 0.3% of the Bardeen-Cooper-Schrieffer²⁰ (BCS) prediction shown by the solid line. From the value for $T_c/\theta_D = 0.0085$, it might be expected to be a weak-coupling superconductor but the closeness of the fit to BCS is probably fortuitous. It may be that strong-coupling effects²¹ are just cancelled by anisotropy effects.²² One of the objects of this work is to study deviations in superconducting characteristics which are brought about by magnetic impurities, so it is convenient to start with a host metal which closely obeys the BCS theory.

Several other characteristics of Th can be deduced from these data. If the low-temperature critical-field results are fit to the BCS equation and if the molar volume v is taken to be 19.7 cc/mole,¹³ then the electronic specific-heat coefficient γ can be derived to be 4.34 mJ/mole °K, compared with the calorimetric value⁹ of 4.31 mJ/mole °K. This then implies a density of states of 1 spin of 0.92 states per eV per molecule. From the slope of the critical-field curve near $T = T_c$, one can derive the jump in specific heat at the superconducting transition $C_n - C_s$ by the thermodynamic

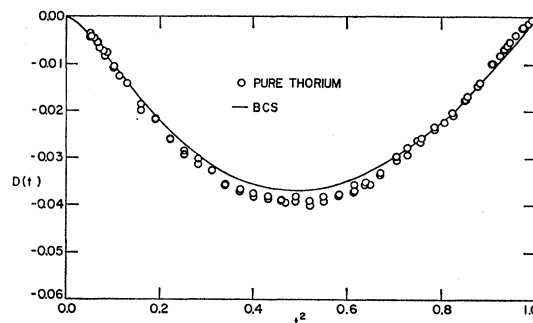


FIG. 5. Critical-field curve of pure Th ($\rho = 0.0128 \mu\Omega$ cm) plotted as deviations from a fiducial parabola through H_0 and T_c .

²⁰ J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

²¹ J. C. Swihart, D. J. Scalapino, and Y. Wada, Phys. Rev. Letters 14, 106 (1965).

²² J. R. Clem, Phys. Rev. 153, 449 (1967).

TABLE II. Characteristics of pure Th and Th-Gd alloys.

| Sample | $\rho(4.2^\circ\text{K})$ $\mu\Omega\text{ cm}$ | T_c $^\circ\text{K}$ | H_0 Oe | $\Gamma/\Delta_p(0)$ | $2\Delta(0)/k_B T_c$ | $(C_s - C_n)/(C_s - C_n)_p$ |
|------------------|--|---------------------------|-------------------|----------------------|----------------------|-----------------------------|
| Pure Th-1200 | 0.0128 | 1.390 ± 0.002 | 159.22 ± 0.10 | 0 | 3.53 | 1.00 |
| Pure Th-35 | 0.45 | 1.364 ± 0.003 | | | | |
| Th 0.10 at. % Gd | 0.63 | 1.110 ± 0.002 | 123.06 | 0.140 | 3.91 | 0.70 |
| Th 0.15 at. % Gd | 0.75 | 0.900 ± 0.002 | 96.16 | 0.238 | 4.30 | 0.52, 0.40 |
| Th 0.20 at. % Gd | 0.86 | 0.761 ± 0.002 | 78.63 | 0.298 | 4.66 | |

relations²³

$$C_n - C_s = (vT_c/4\pi) (dH_c/dT)_{T_c}^2. \quad (1)$$

The magnetic data give a jump of 9.0 mJ/mole $^\circ\text{K}$ compared with the calorimetric value⁹ of 8.4 mJ/mole $^\circ\text{K}$. Another manifestation of the close agreement with BCS is that 9.0 mJ/mole $^\circ\text{K}$ is $1.5 \gamma T_c$ compared with the theoretical value of $1.43 \gamma T_c$.

Within BCS the free energy at $T=0$ is related to the energy gap by²⁰

$$2\Delta(0) = 2H_0[\pi k_B^2 v/6\gamma]^{1/2}, \quad (2)$$

and hence the experimental values of H_0 , γ , and v would imply an energy gap for thorium of $3.53 kT_c$. Not only is the shape of critical-field curve on a reduced scale BCS-like but the magnitude of H_0 gives the BCS value of energy gap. The characteristics of pure Th have been summarized in Table II.

Alloys

When small amounts of Gd are added to pure Th, the transitions broaden slightly and hysteresis becomes

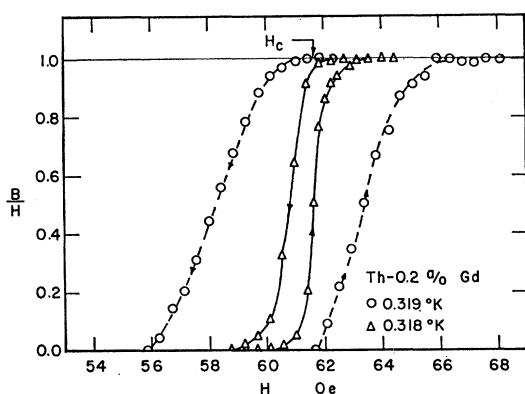


FIG. 6. Magnetization curves for Th 0.20 at. % Gd. The dashed curve was measured after an anneal of $\frac{1}{2}$ h at 800°C . The solid curve was measured after a subsequent anneal of 7 days at 1200°C . H_c was determined from the average of the field-increasing and the field-decreasing transitions as extrapolated to $B/H=1.0$.

²³ D. Shoenberg, *Superconductivity* (Cambridge University Press, Cambridge, England, 1952).

a relevant factor. The general character of this hysteresis is qualitatively different from the supercooling in pure Th in that there is no catastrophic drop in magnetization after nucleation takes place and the sample exhibits hysteresis loops anywhere in the intermediate state. For the Th 0.2 at. % Gd sample, a rather thorough study has been made of the hysteresis and the results are briefly illustrated in Fig. 6. After an anneal at 800°C for $\frac{1}{2}$ h the transitions were rather broad and the hysteresis was about 8.0% of H_c or 5.0 Oe at 0.3°K , as shown by the dashed line of Fig. 6. After a second anneal of 7 days at 1200°C , the transitions at the same temperature were sharpened and the hysteresis decreased substantially, as shown by the solid line of Fig. 6. Even though the superconducting-to-normal transition is only 0.50 Oe wide, it still is approximately twice that to be expected from the demagnetizing factor so further annealing might yet improve matters. No further anneal was attempted, however, because 0.6% hysteresis was adequate for the purpose of these experiments. Both the field-increasing and the field-decreasing transitions were shifted toward

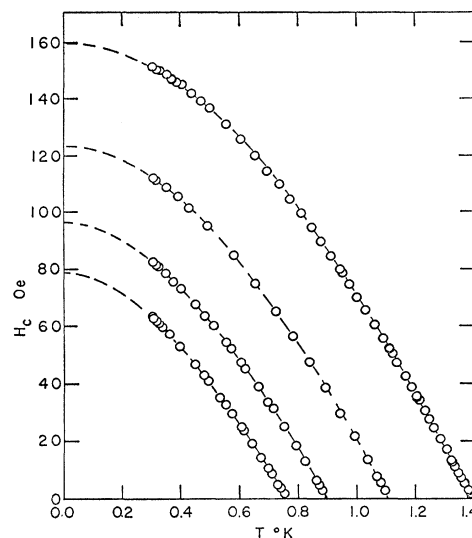


FIG. 7. Critical-field curves for the Th-Gd alloys. The dashed curves were calculated from AG using the ratio of T_c/T_{cp} to determine Γ .

the center by the annealing and on the basis of this fact we had defined the critical field H_c to be the midpoint, as shown by the arrow in Fig. 6. Many features of the hysteresis in these alloys are similar to results for pure Pb reported by Decker *et al.*²⁴

The absence of a well-defined nucleation field precludes a direct measure of κ for these alloys but an estimate can be made of the normal-state electrical resistivities through the relation²⁵ $\kappa = \kappa_0 + 7.53 \times 10^3 \rho \gamma^{1/2}$, where ρ is in $\mu\Omega$ cm and γ is in erg/cc. For this alloy system, the electrical resistivity increases by about $2.5 \mu\Omega$ cm/at.% Gd, so κ would increase from 0.4 for

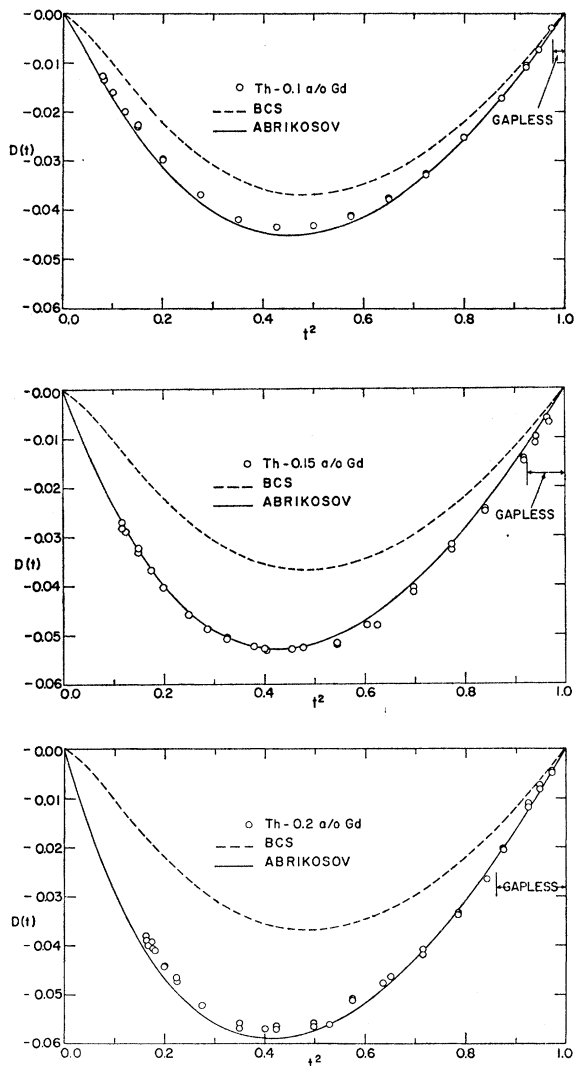


FIG. 8. Deviation functions for the Th-Gd alloys.

²⁴ D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. **112**, 1888 (1958).

²⁵ B. B. Goodman, IBM J. Res. Develop. **6**, 63 (1962); Phys. Rev. Letters **6**, 597 (1961).

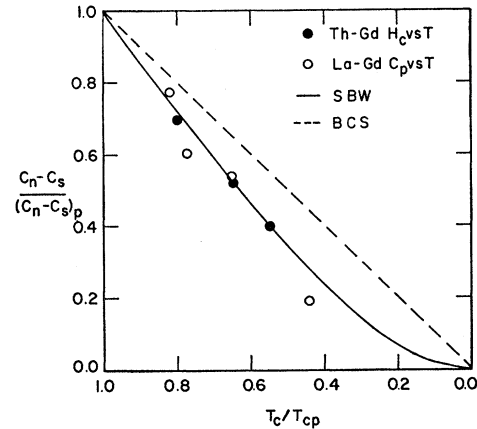


FIG. 9. The jump in specific heat at T_c . Gd concentration increases to the right.

pure Th to 0.7 for the 0.2% Gd sample. Higher concentrations are probably type-II.

There are at least two different scattering times which are important in determining the behavior of these alloys. The transport scattering time (τ_t), which includes both magnetic and nonmagnetic processes, determines the normal-state resistivity and hence determines whether the material is a type-I or a type-II superconductor. The magnetic or spin-flip scattering time (τ_s) determines the depression of the superconducting transition temperature.¹ From the normal-state resistance of $1\mu\Omega$ cm, it would be expected that τ_t is of the order of 10^{-14} sec, and from the depression of T_c it would be expected that $\tau_s = \hbar/\Gamma$ is approximately 10^{-11} sec. Hence for this system the magnetic processes are a very small fraction of the total scattering.

The critical-field data for these alloys are shown in Fig. 7. As T_c/T_{cp} decreases, the curves remain nearly parabolic in shape but the ratio of H_0/T_c steadily decreases. The theoretical curves shown by the dashed lines were calculated from AG as described earlier. Once the properties of pure Th are known, one additional parameter T_c/T_{cp} is sufficient to determine Γ and the critical-field curve at all temperatures.

If the data are cast as deviations from a fiducial parabola through the measured value of T_c and the AG value of H_0 as shown in Fig. 8, then it can be seen that the deviations become more negative with increasing Gd, just as the theory predicts. When one considers that the only adjustable parameter here is T_c , these data must be taken as very strong quantitative support for the theory.

An alternative but related way to compare the data with the theory is to calculate the specific-heat jump at T_c by Eq. (1). These results are shown by the solid dots of Fig. 9. Note that the abscissa shows T_c/T_{cp} decreasing to the right, corresponding to Γ or impurity concentration increasing to the right. If the

impurities were nonmagnetic, then BCS should apply and the specific-heat jump would be proportional to T_c/T_{cp} , as shown by the dashed line. For magnetic impurities, SBW predicts the solid curve, in excellent agreement with the experiments. The open circles are calorimetric determinations of the specific-heat jump take in this laboratory for La-Gd alloys.^{6,26} Both the specific-heat and critical-field results show the effect but the critical-field data illustrate the point to much higher accuracy.

Values of ρ , H_0 , T_c , $\Gamma/\Delta_p(0)$, and $C_s - C_n|_{T=T_c}$ are all summarized in Table II. A complete tabulation of all the data is presented elsewhere.¹³

CONCLUSIONS

Pure thorium is a weak-coupling type-I superconductor with critical-field curves which follow the BCS

²⁶The data for the highest concentration have not yet been published in Ref. 6.

theory to an accuracy of 0.3%. For additions of Gd impurity in concentrations up to 0.2%, the samples remain type-I superconductors which exhibit a Meissner effect in the gapless regime near T_c as well as the regime where there is a well-defined gap at low temperatures. The AG-SBW theory^{1,12} predicts the critical-field curves for Th-Gd alloys to an accuracy of better than 0.5%.

ACKNOWLEDGMENTS

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Radiative Decay of Coulomb-Stimulated Plasmons in Spheres*

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We have studied the excitation of plasmons by energetic electrons, and their subsequent decay, in small spheres. We estimate that plasma decay light from electron bombardment of small, randomly distributed metal spheres suspended in a dielectric medium may be emitted with intensity comparable with that found in experiments on plasma decay light from metallic slabs.

I. INTRODUCTION

THE characteristic absorption of light by small metal spheres has recently been studied experimentally by Doremus¹ and theoretically by Kawabata and Kubo.² Doremus's data for the wavelength dependence of the absorption coefficient for 100 Å silver and gold spheres show peaks which are explained on the basis of plasma resonance absorption by the conduction electrons. Kawabata and Kubo have used linear response theory to explain the widths of these resonances in terms of the damping of the plasma oscillations by transfer of energy to single-electron modes. They also

take account of radiation damping, and their results show good agreement with Doremus's data.

The successful explanation of light absorption peaks in terms of collective resonances in a spherical electron gas leads one to speculate about the possibility of stimulating similar resonances by means of energetic, nonrelativistic electrons and observing the radiative decay of these collective states. It is well known that such resonances can be produced in metal foils by electron bombardment and that light from the damping of these resonances can be observed.³ Plasmon decay light from foils was first predicted by Ferrell.⁴ Recent characteristic energy-loss experiments by Fujimoto,

* Research sponsored by the U.S. Atomic Energy Commission under contract with Union Carbide Corporation.

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