Adiabatic Magnetization of a Superconducting Sphere

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Measurements are reported of the temperature changes accompanying suppression of the superconductivity of a lead sphere by adiabatic magnetization. Observations were made in the temperature range 2.1°K to 4.3°K for magnetizations into the complete range of the intermediate state. The magnetic fields were applied as step functions of time, and allowance was made for the resulting eddy-current heating. After magnetization, practically complete temperature equilibrium was established in the order of 15 seconds. Temperature changes observed on magnetization were in quantitative agreement with values predicted from calorimetric data and demonstrate a linear relationship between the applied magnetic field and the fraction of normal metal produced in the intermediate state.

INTRODUCTION

AT a given temperature, the molar entropy of a metal in the superconducting state is smaller than the molar entropy of the metal in the normal state produced by the application of a magnetic field in excess of the critical field at that temperature. This entropy difference accrues only during increase of the magnetic field from a threshold-field value determined by specimen geometry to the critical-field value for bulk material.[1] The manner of variation of the entropy during this intermediate state transition has received theoretical attention.[2] Thus, utilizing specific heat and critical-field data, one can predict the cooling to occur upon the isentropic magnetization of a superconducting sphere. It has been observed by other investigators[3–5] that cooling of a superconductor results from the application of a magnetic field sufficient to produce the intermediate state. However, experimental verification of the predicted cooling was not accomplished. This paper is a report of an experiment designed to measure the temperature changes induced in a superconducting sphere by adiabatic magnetizations and to test the predicted linear relationship between the applied-magnetic field (H) and the fraction (x) of normal metal produced in the intermediate state; *viz.*, $x=(3H/H_c-2)$, where H_c is the critical-magnetic field for bulk material at the specimen temperature.

EXPERIMENTAL PROCEDURE

Measurements were taken of the cooling produced by adiabatic magnetization of a superconducting lead sphere. Observations were made in the temperature range 2.1°K to 4.3°K for fields in the range $0<h\leq 1.03$, where h is the ratio of applied field to critical-magnetic field. This range of applied fields corresponds to $0<x\leq 1$, where x is the fraction of the sphere that becomes normal when the sphere is subjected to a magnetic field sufficient to produce the intermediate state, *viz.* $\frac{2}{3}<h<1$. A carbon thermometer[6] was employed to measure temperature, the resistance of the thermometer being automatically recorded by a system having a response time of about five seconds. Temperature changes could be measured to approximately ± 0.005°K. Practically complete equilibrium was reached in approximately 15 seconds after the application of the magnetic field. The fact that equilibrium was established in such a short time is in agreement with previous observations[7,8] and indicative of good thermal contact between the thermometer and the specimen. Figure 1 presents a typical record obtained on magnetization.

Since the conditions for reversibility were difficult to realize, changes in magnetic field were applied as step functions of time, and allowance was made for induced eddy-current heating. The eddy currents induced in the sphere were calculated and the excess magnetic energy was used, together with the isentropic cooling predicted by calorimetric data, to calculate the expected theoretical temperature change. The experimental conditions of measurement were the same as those reported in a previous publication[9] except that since the sample temperature changed, the magnetizations were adiabatic rather than isothermal. Thus, the eddy-current heating was calculated by the method previously outlined,[9] but an ideal isentropic-magnetization curve was used rather than an ideal isothermal-magnetization curve. In these

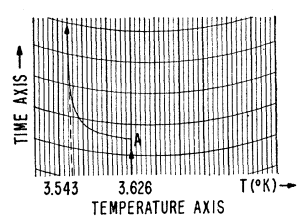

FIG. 1. Typical temperature record obtained on magnetization of a superconducting sphere. Markers on the time axis are at 15-second intervals. The magnetic field was turned on at A. Intervals along the temperature axis are only approximately equal as thermometer resistance is recorded. This record is of trial No. 43 of Table II.

[1] R. L. Dolecek, Phys. Rev. **82**, 102 (1951).
[2] D. Shoenberg, *Superconductivity* (Cambridge University Press, London, 1938), Chap. V; see also second edition (1952), Chap. III.
[3] K. Mendelssohn and J. R. Moore, Nature **133**, 413 (1934).
[4] W. H. Keesom and J. A. Kok, Physica **1**, 595 (1934).
[5] Mendelssohn, Daunt, and Pontius at Congres Internacionale du Froid, Le Haye, 1936 (unpublished).
[6] J. R. Clement and E. H. Quinnell, Rev. Sci. Instr. **23**, 213 (1952).
[7] D. Shoenberg, Proc. Cambridge Phil. Soc. **33**, 559 (1937).
[8] W. H. Keesom and P. Van Laer, Physica **3**, 173 (1936).
[9] R. L. Dolecek, Phys. Rev. **94**, 540 (1954).

TABLE I. Magnetization measurements at various initial temperatures.

Trial No.	Initial temperature °K	Applied h	Computed eddy-current heating °K	Computed isentropic cooling °K	Theoretical temperature change expected °K	Experimental temperature change observed °K	Deviation °K
35	4.378	1.02	0.084	0.256	−0.172	−0.174	−0.002
36	3.988	0.98	0.085	0.275	−0.190	−0.187	+0.003
37	3.755	0.95	0.094	0.287	−0.193	−0.186	+0.007
56	3.625	0.93	0.100	0.280	−0.180	−0.179	+0.001
61	3.241	0.97	0.213	0.382	−0.169	−0.179	−0.010
60	2.937	0.97	0.315	0.442	−0.127	−0.153	−0.026
59	2.670	1.03	0.555	0.579	−0.024	−0.078	−0.054
58	2.057	1.02	1.074	0.707	+0.367	+0.262	−0.105

calculations the critical-field data of Daunt *et al.*[10] were used and it was assumed that the critical-field curve was parabolic. The isentropic temperature changes for magnetization were computed using specific-heat data for lead[11–13] adjusted to yield, at the transition temperature, zero entropy difference between the superconducting and normal states. Also used were some recent measurements[14] of the atomic heat and latent heat of transition of superconducting lead which indicate a quartic dependence of the critical-magnetic field on temperature.[9] Since the Debye temperature of lead is not constant in the temperature interval studied, a lattice atomic heat of the form AT^3+BT^5 was employed.[15] The equations assumed for the entropies were

Molar entropy of superconducting lead

$$=(1.377T^3+0.0118T^5+1.01\times10^{-6}T^7)\times10^{-4}\ \text{cal}/°\text{K},$$

Molar entropy of normal lead

$$=(8.0T+1.267T^3+0.011T^5)\times10^{-4}\ \text{cal}/°\text{K}.$$

These equations yield entropy differences between the superconducting and normal states in good agreement with the values obtained by Daunt and Mendelssohn.[10]

EXPERIMENTAL RESULTS

In Table I typical measurements made on magnetizations from zero magnetic field to h, computed at the final temperature, are given together with temperature changes resulting from eddy-current heating and computed values of theoretical temperature changes expected. The value of the eddy-current heating ranges from a small to a predominant part of the expected theoretical cooling. Thus, the measurements test not only the accuracy of the eddy-current heating calculations but also the correctness of interpretation of the behavior of a superconducting sphere in the intermediate state and the accuracy of the functions assumed for the entropy and magnetic behavior of lead. The deviations encountered in these observations are quite large in absolute value and, at the lower temperatures, there appears to be systematic discrepancy. However, in all instances the deviation is less than ten percent of the sum of the calculated temperature changes involved. In the computation of the theoretical temperature change expected, experimental data for specific heats and critical-magnetic fields were employed, the magnetization curve for a perfect sphere was used, and no allowance was made for hysteresis effects. In view of these limitations, the data obtained are in good agreement with theory and verify (within 10 percent) the current interpretation of the behavior of a superconducting sphere in the intermediate state.

The predicted linear relationship between the applied magnetic field and the fraction of normal metal produced in the intermediate state can be tested by a calorimetric experiment in which magnetizations throughout the range of the intermediate state are all performed from a fixed initial temperature. The energy per mole absorbed by a conversion of superconducting metal to the normal state is given by the latent heat Q multiplied by the fraction converted dx, and this under adiabatic conditions will result in an absorption of heat energy of CdT from the metal, where C is the molar specific heat. If the magnetizations are all performed from the same initial temperature, then $Q/C\simeq$constant and one would expect $dT/dx\simeq$constant. Thus, if the fraction (x) converted is linearly related to the applied field H, a plot of temperature change resulting from magnetization *versus* x calculated from the magnitude of the applied magnetic field should yield a straight line of slope Q/C.

From an initial temperature of 3.62±0.02°K, measurements were taken of the cooling produced by adiabatic magnetization into the complete range of the intermediate state. The measurements are given in Table II. Except for the small temperature changes near $x=0$, the deviations again are generally less than ten percent of the temperature changes involved. The eddy-current corrections in Table II include a small vibration heating incurred upon the sudden application of the magnetic field to the spherical specimen. This correction

[10] J. G. Daunt and K. Mendelssohn, Proc. Roy. Soc. (London) **160**, 127 (1937); Daunt, Horseman, and Mendelssohn, Phil. Mag. **27**, 754 (1939).
[11] W. H. Keesom and J. N. van den Ende, Leiden Comm. **19**, No. 213C (1931).
[12] J. R. Clement and E. H. Quinnell, Phys. Rev. **85**, 502 (1952).
[13] Horowitz, Silvidi, Malaker, and Daunt, Phys. Rev. **88**, 1182 (1952).
[14] These measurements, performed at the Naval Research Laboratory, are in preparation for publication.
[15] M. Blackman, Proc. Roy. Soc. (London) **A159**, 416 (1937).

TABLE II. Magnetization measurements at fixed initial temperatures.

Trial No.	Initial temperature °K	$x=\left(\frac{3H}{H_c}-2\right)$	Computed eddy current heating °K	Computed isentropic cooling °K	Theoretical cooling computed °K	Experimental cooling observed °K	Deviation °K
49	3.633	0.024	0.015	0.008	−0.007	0.002	−0.009
47	3.595	0.068	0.015	0.023	0.008	0.008	0.00
48	3.641	0.090	0.015	0.030	0.015	0.013	+0.002
51	3.633	0.135	0.015	0.046	0.031	0.028	+0.003
44	3.618	0.182	0.015	0.064	0.049	0.050	−0.001
53	3.624	0.238	0.015	0.084	0.069	0.065	+0.004
43	3.626	0.298	0.017	0.106	0.089	0.083	+0.006
54	3.624	0.400	0.026	0.143	0.117	0.110	+0.007
42	3.622	0.497	0.040	0.178	0.138	0.132	+0.006
55	3.621	0.565	0.050	0.201	0.151	0.139	+0.012
41	3.621	0.677	0.072	0.242	0.170	0.166	+0.004
56	3.625	0.786	0.100	0.280	0.180	0.179	+0.001
40	3.588	0.823	0.116	0.299	0.183	0.187	−0.004
57	3.630	0.893	0.132	0.315	0.185	0.183	+0.002
39	3.654	1.00	0.171	0.354	0.183	0.192	−0.009

is a predominant part of the eddy-current correction near $x=0$. In the computation of the temperature changes, experimental data for specific heats and critical-magnetic fields were employed, the magnetization curve for a perfect sphere was used, and no allowance was made for hysteresis effects. In view of these limitations, the data obtained are in good agreement with theory and verify (within 10 percent) the linear dependence of the fraction of the sphere in the normal state upon the average magnetic induction of the whole specimen. For test of the predicted linear dependence, the fraction (x) computed from the value of magnetic field applied is to be plotted against the total temperature change which is obtained by summing the eddy-current heating and the observed experimental cooling. This plot is presented in Fig. 2.

Except near the ends, the relationship is quite linear and since the fraction (x) was computed from the value of the applied magnetic field, the agreement substantiates the assumption that the fraction (x) of normal metal produced in the intermediate state of a superconducting sphere is a linear function of the applied magnetic field. This confirms the observations of Keesom and Van Laer[16] who have demonstrated the linear dependence for a tin ellipsoid having a demagnetizing coefficient of 0.056. That departures occur near the ends of the curve may be related to the fact that, in the calculations, an ideal magnetization curve was assumed for the sphere with no discontinuities on entering or leaving the intermediate state and no hysteresis. Variation of the latent and specific heat might produce the deviation from linearity at the larger values of x.

The slope of the curve for $x<0.6$, where eddy-current corrections are negligible, is about 0.34. As previously shown, this slope should equal Q/C. From calorimetric data one calculates an expected value of $Q/C\simeq 0.32$. This is in reasonably good agreement with the slope of the curve.

The effect of eddy-current heating was nominal for magnetizations involving $h<0.9$. However, it should be pointed out that there is an inversion at a temperature of about one-third the superconducting transition temperature and that below this temperature the eddy-current heating accompanying magnetization by the abrupt application of a magnetic field will exceed the isentropic cooling. See, for example, the eddy-current heating incurred for point No. 58 of Table I. It can be seen that at low temperatures the eddy-current heating of metals under magnetization (and demagnetization) can be of considerable magnitude and should be recognized as a possible source of serious heating. Magnetic cooling cycles should be designed to minimize the effect.

To realize most of the available cooling one may minimize eddy-current heating either by reducing the demagnetization factor of the sample or by applying the magnetic field slowly. For a lead sphere at 3.6°K it was found possible to obtain ninety percent of the theoretical isentropic cooling by traversing the intermediate state in about three minutes.

CONCLUSIONS

Measurements were performed on the adiabatic magnetization of a superconducting sphere, and the

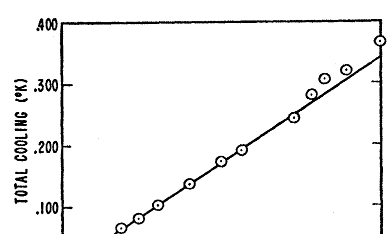

FIG. 2. Demonstration of the linear relationship between the applied magnetic field and the fraction of normal metal produced in the intermediate state. The straight line is drawn with a slope to fit best the data $x\leq 0.6$ for which eddy-current corrections are small.

[16] W. H. Keesom and P. H. Van Laer, Physica 4, 487 (1937).

results confirm the gross aspects of current interpretation of the behavior of a superconducting sphere in the intermediate state. Temperature changes observed on magnetization were in quantitative agreement with values predicted from calorimetric data and demonstrate a linear relationship between the applied magnetic field and the fraction of normal metal produced in the intermediate state.

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Electrical Properties of Silicon Containing Arsenic and Boron

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Electrical conductivity and Hall effect have been measured from 10° to 1100° Kelvin on single-crystal silicon containing arsenic and boron. Extrinsic carrier concentration is computed from Hall coefficient. Analysis of extrinsic carrier concentration indicates the ionization energy of arsenic donor levels to be 0.049 ev and of boron acceptor levels to be 0.045 ev for low impurity concentrations. Fermi degeneracy is found to occur in the range 10^{18} to 10^{19} cm^{-3} impurity concentration. Extrinsic Hall mobility is computed from Hall coefficient and conductivity. Curves of Hall mobility against resistivity at 300°K are computed from theory and compared with experiment. The temperature dependence of lattice-scattering mobility is found from conductivity to be $T^{-2.6}$ for electrons and $T^{-2.3}$ for holes. From conductivity mobility and intrinsic conductivity, it is found that carrier concentration at any temperature below 700°K is given by the expression: $np=1.5\times10^{33}T^3\exp(-1.21/kT)$. The temperature dependence of the ratio Hall mobility/conductivity mobility is determined for holes and electrons.

1. INTRODUCTION

An extensive investigation of the fundamental electrical properties of silicon has not been published since 1949, when Pearson and Bardeen reported on silicon containing boron and phosphorus.[1] Their experimental work was necessarily limited because neither single crystals nor the means for measuring below 77°K were then available. Recently, it seemed opportune to reinvestigate silicon: good quality single crystals were available; it became possible to make the low temperature measurements necessary to locate precisely the impurity levels; there was considerable interest in silicon and fundamental information was needed.

Electrical conductivity and Hall effect have been measured from 10° to 1100°K on single crystals of silicon containing arsenic and boron. Unfortunately a complete analysis of the data is not possible at present. For example, one needs to know more about the scattering processes which produce the departure from the $T^{-1.5}$ law of lattice scattering mobility found for both holes and electrons; it is shown that electron-hole scattering is probably negligible below 1200°K; only a very rough estimate of optical mode scattering can be made; the possibility of scattering by multiple constant energy surfaces and by band splitting due to spin-orbit coupling was suggested by C. Herring but is neglected because theory is lacking. The meaning of the low temperature Hall effect in some samples is uncertain. Behavior of computed Hall mobility suggests the presence of impurity level conduction for which theory is unavailable. Determination of the mass parameter by fitting carrier concentration data in the extrinsic range is faced with two new problems for which theory is unavailable: the apparent dependence of the mass parameter on impurity concentration and the possible existence of excited states associated with the impurity atoms.

[1] G. L. Pearson and J. Bardeen, Phys. Rev. **75**, 865–883 (1949).

1.1 Methods

Crystals were prepared by E. Shannon of these laboratories using the Teal-Little pull technique.[2] In each case two crystals were pulled from du Pont silicon and then combined with the added impurity in a melt from which the final crystal was pulled. Samples were bridge shape[3] cut with the [110] direction along the length of the sample. The samples were sandblasted. Contact was made to n-type samples by bonding through gold plate with Sb-doped gold wire. Contacts to p-type samples were pressure contacts on rhodium plate.

1.2 Symbols

The symbols used have been defined previously by the authors.[4]

2. EXPERIMENTAL RESULTS

Conductivity and Hall coefficient in the intrinsic range of silicon are shown in Fig. 1. The Hall coefficient

[2] G. K. Teal, Phys. Rev. **78**, 647 (1950); Teal, Sparks, and Buehler, Phys. Rev. **81**, 637 (1951).
[3] P. Debye and E. Conwell, Phys. Rev. **93**, 693–706 (1954).
[4] F. J. Morin and J. P. Maita, Phys. Rev. **94**, 1525 (1954). See Sec. 3 for definition of symbols.