SURFACE NUCLEATION FIELD H_{c3} OF PURE LEAD

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New measurements of $H_{c3}(T)$ are presented for pure Pb. The analysis of the present data suggests that $H_{c3}(T)$ in pure Pb depends strongly upon the type of electron scattering at the surface. When this effect is properly taken into consideration, it is possible to derive the bulk critical field $H_{c2}(T)$ and Ginzburg-Landau parameter $\kappa_1(T)$ of pure Pb, with results very close to predictions from recent theories of strong-coupling superconductors.

There is a renewed theoretical interest in reliable $H_{c3}(T)$ data for pure Pb since there have been recent papers concerned with the influence on $H_{c3}(T)$ of strong-coupling effects[1,2] and of the scattering mechanism of the electrons at the surface of a superconductor.[3] Both theories of strong-coupling effects[1,2] predict that the slope dH_{c3}/dT at $T=T_c$ should be substantially less than the value of about 206 Oe/°K which one deduces from available experimental data.[4] The theory of Lüders[3] predicts that the degree of diffuse electron scattering at the surface of a clean superconductor affects $H_{c3}(T)$ at all temperatures but does not affect the slope dH_{c3}/dT at T_c. In view of this it is possible to determine this slope from experiments even when one can measure $H_{c3}(T)$ only at temperatures below which $H_{c3}>H_c$.

$H_{c3}(T)$ data were so far obtained from surface resistance,[4-6] from magnetization,[7,8] and from tunneling characteristics.[9] Between H_c and H_{c3} the magnetic response is diamagnetic or paramagnetic according to whether the applied field H is increasing or decreasing.[5] As one approaches H_{c3} the order parameter becomes arbitrarily small and demagnetization plays no part. Geometry, however, remains important: The surface sheath will develop only on surfaces which are parallel to the applied field.[10] Near H_{c3} the magnetization of a clean type-I superconductor[11] is proportional to $(H_{c3}-H)^2$. Our measurements indicate that near H_{c3} the surface resistance[12] also varies like $(H_{c3}-H)^2$. This behavior is very different from the one obtaining for dirty type-II superconductors. For these both magnetization[13] and surface resistance[14] vary linearly, i.e., $\propto H_{c3}-H$ near H_{c3}, and the determination of H_{c3} is generally less ambiguous. Magnetization is a volume effect so that near H_{c3}, magnetization measurements are not very sensitive. For surface resistance the sensitivity remains high because normal-state skin depth, anomalous for pure Pb near 0°K, and sheath thickness, equal to coherence length[15] ξ near H_{c3}, are of similar size. Consequently we have chosen surface resistance to determine H_{c3}.

Several factors are known to produce anomalous values of H_{c3}: (1) an increase of κ near the surface because of imperfections of one sort or another, (2) surface roughness with asperities of dimensions larger than ξ,[16] and (3) the presence of a dielectric at the surface.[17] Our aim is to avoid these causes of anomalous H_{c3} but at the same time we want to vary the proportion p of electrons that are scattered diffusely at the surface. We can summarize our observations as follows. Pure Pb freshly evaporated on room-temperature substrates is extremely compact, yielding mean free paths of more than 1 μ and extremely smooth surfaces[18] (our evaporated samples are about 5 μ thick). The same holds for rolled, annealed, and electropolished samples. As soon as they come in contact with atmosphere these samples oxidize somewhat and we believe that this produces the proportion $p\cong 0.5$ of diffuse surface scattering that our $H_{c3}(T)$ data suggest for samples exposed to atomosphere for about 15 min. However, none of the three effects listed is detected. Effect (3) is seen with samples exposed to atmosphere for a few hours or days.[19] To increase p and avoid effect (2) we polish both types of samples with a polishing powder of 500-Å grit size. If the samples are measured immediately after polishing we observe effect (1), but when the samples are stored under high vacuum for a few days we find that effect (1) disappears. This indicates that pure Pb anneals at room temperature. Effect (2) is observed when the samples are polished with powder of a coarser grit or are sandblasted. The polished samples are mirrorlike in appearance but yield $H_{c3}(T)$ data consistent with completely diffuse surface scattering, i.e., $p=1$.

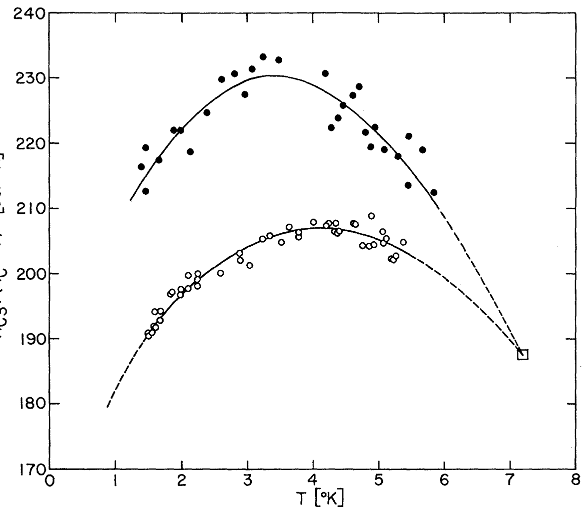

FIG. 1. Plot of the ratio $H_{c3}/(T_c-T)$ vs T for two samples of pure Pb. Open circles, freshly evaporated or electropolished, $p=0.48$; solid circles, mechanically polished (500-Å grit size), $p=1$; square, extrapolation to T_c as described in text.

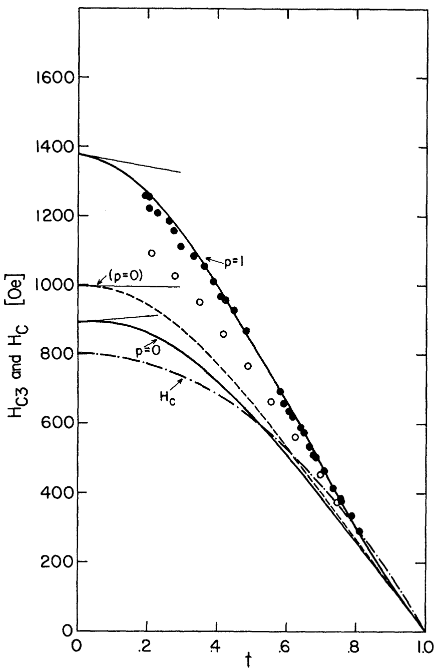

FIG. 2. H_{c3}, H_{c2}, and H_c (Ref. 23) versus reduced temperature t. The solid curves are upper and lower limits for H_{c3} when $p=1$ and $p=0$. The curve labeled ($p=0$) corresponds to $p=0$ in Eq. (2), i.e., $H_{c3}(t) = 1.695 H_{c2}(t)$. The data points are as in Fig. 1.

Details of the experimental technique are given elsewhere,[18] but we want to mention here that our dc field was parallel to the sample surface within $\pm\frac{1}{10}$ deg and was also parallel to the microwave electric field.

When $p>0$ Lüders's theory[3] predicts that near T_c the slope of $H_{c3}(T)$ should increase as T decreases. To check this prediction we plot in Fig. 1 the quantity $H_{c3}/(T_c-T)$ against T. We find that this quantity goes through a maximum indicating that indeed $H_{c3}(T)$ is concave near T_c. We note in Fig. 1 that the data relating to freshly prepared samples show less scatter than those from the polished samples. We shall, therefore, look at the data from the freshly prepared samples in more detail. First we try to find the slope of $H_{c3}(T)$ at T_c. To this end we plot H_{c3} against the function $f(t) = (1-t^2)/(1+t^2)$ and find that $H_{c3}(f)$ deviates little from a straight line (t = reduced temperature). Extrapolating toward $f=0$ and $f=1$ yields the two important parameters $H_{c3}(T=0) = 1181 \pm 4$ Oe and $dH_{c3}/dT = -187.5 \pm 2$ Oe/°K at T_c. Now we know that the second of these parameters is independent of p, and this is borne out in Fig. 2. $H_{c3}(0)$, however, depends strongly on p.

If we were to analyze our data assuming the ratio H_{c3}/H_{c2} to be a constant, independent of temperature, we would have a critical field $H_{c2}(T)$ which, like our $H_{c3}(T)$, has a concave curvature near T_c. This seems to us rather implausible. There has been a large body of evidence[20] in the case of dirty superconductors favoring a constant ratio $H_{c3}/H_{c2} = 1.695$. However, our data pertain to the clean limit and it is only near T_c that there is good ground for assuming $H_{c3}/H_{c2} = 1.695$, independent of the strong coupling.[1,2] To find $H_{c2}(T)$, then, we can divide $H_{c3}(T)$ by 1.695 near T_c and extend the data analytically to lower temperatures with any good theory for $H_{c2}(T)$. However, there is not, at present, a good theory of $H_{c2}(T)$ in the clean strong-coupling limit. In view of this we have used a different approach. It is known in the weak-coupling limit that $H_{c2}(T)$ is linear with T down to particularly low temperatures when the system is clean rather than dirty,[21] and in an application to niobium it has been shown[22] that strong-coupling effects alone did not change $H_{c2}(T)$ by more than 2%. Excluding the unlikely possibility that effects connected with the Fermi surface of Pb strongly distort $H_{c2}(T)$ near T_c, we assume $H_{c2}(T)$ to be linear down to $T = 5$°K. We then explain the concave behavior of $H_{c3}(T)$ by means of a proportion p of diffuse electron scattering at the surface. Lüders[3] has derived the following formulas:

$$H_{c3}(t) = 1.695F(t)H_{c2}(t), \tag{1}$$

where

$$F(t) = 1 + 0.382p(1-t)^{1/2} \text{ for } p > 0, \tag{2}$$

and

$$F(t) = 1 - 0.105(1-t) \text{ when } p = 0. \tag{3}$$

Formula (2) is valid for $p > 0$ and not too small.[3] Equations (1) to (3) have been derived from generalized and linearized Ginzburg-Landau equations containing terms with spatial derivatives up to fourth order. While these equations are certainly not valid down to $t = 0$, they remain valid near H_{c3} to much lower temperatures than the ordinary Ginzburg-Landau equations. Our data suggest, in fact, that Lüders's formulas are reasonably good down to at least 1.5°K. Lüders's theory does not consider strong-coupling effects, but this does not seem noticeably to impair its validity. This may be so because near T_c a form of Ginzburg-Landau equations can also be derived for strong-coupling superconductors,[2] and these may in turn be generalized to lower temperatures.

Combining Eqs. (1) and (2) we obtain a function $H_{c3}(t)$ which is concave in the region above 5°K. If we require the slope of $H_{c3}(T)$ at 5°K to be given by our data we deduce $p = 0.48$. With this value of p and assuming Eqs. (1) and (2) to hold at all temperatures we calculate $H_{c2}(T)$ as given in Table I. We can also calculate $H_{c3}(T)$ as it would obtain for $p = 1$ and $p = 0$ with Eqs. (2) and (3). It is satisfying to see in Fig. 2 that the data from the polished samples very closely lie on the curve for $p = 1$. Since there is no adjustment anywhere to obtain this agreement we consider it as a justification of Lüders's theory[3] to temperatures as low as 1.5°K. The limitations of this theory at very low temperatures are evident in Fig. 2, where it is seen to yield for $p = 0$ a positive slope of $H_{c3}(T)$ at $T = 0$ and for $p > 0$ a negative such slope. At $t = 1$ all curves of $H_{c3}(t)$ merge together with the same slope $dH_{c3}/dT = -187.5 \pm 2$ Oe/°K, a value which is in good agreement with recent theories of strong-coupling superconductors.[1,2] Depending on the value of p one should observe $H_{c3}(T') = H_c(T')$, that is the disappearance of surface superconductivity, at very different temperatures T'. For $p = 0$ we predict $T' = 3.75$ °K and for $p = 1$, $T' = 5.88$°K. This implies in particular that it is possible at 4.2°K, if p is sufficiently low, for surface superconductivity not to develop on pure Pb surfaces. Finally we have been able to deduce what we believe to be reliable values of $H_{c2}(t)$ and $\kappa_1(t)$ for pure Pb. These are listed in Table I. The temperature dependence of $H_{c2}(t)$ is close to that predicted by Helfand and Werthamer[21] in the weak-coupling limit for a metal with spherical Fermi surface. Our ratio $h_{c2}(t) = H_{c2}(t)/H_{c2}(0)$ deviates by less than 0.02 from both the clean and dirty limits of their theory, and has at $t = 1$ a slope of 1.341 compared with 1.375 in the clean and 1.444 in the dirty limits. Whereas our $h_{c2}(t)$ is below theirs for $t > 0.6$, it is larger when $t < 0.6$. This behavior of $H_{c2}(t)$ combines with the particular $H_c(t)$ of Pb[23] to give a strongly temperature dependent $\kappa_1(t)$ as reported in Table I.

Table I. Collected data of the present investigation.

T (°K)	H_{c2} (Oe)	$H_{c2}(t)/H_{c2}(0)$	$\kappa_1(t)$	$\kappa_1(t)/\kappa(1)$
0	594[a]	1	0.523	1.595
1.50	554	0.933	0.508	1.55
2.00	524	0.882	0.496	1.51
2.50	486	0.818	0.481	1.465
3.00	444	0.747	0.464	1.415
3.50	398	0.670	0.449	1.37
4.00	349	0.588	0.432	1.32
4.50	296	0.498	0.414	1.26
5.00	241	0.406	0.395	1.205
5.50	186	0.313	0.376	1.145
6.00	131	0.221	0.359	1.095
6.50	76.5	0.129	0.347	1.06
7.00	22	0.037	0.333	1.015
7.20	0	0	0.328	1

[a] Extrapolated value as explained in text for $H_{c3}(T)$.

A preliminary investigation of niobium reveals a similar behavior in this material. However, polishing introduces some strain at the surface of the samples, i.e., an increase of κ_1 toward the surface, which does not anneal at room temperature. A detailed account will be given when more data have been collected.

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[1] G. Eilenberger and V. Ambegaokar, Phys. Rev. 158, 332 (1967).

[2] E. D. Yorke and A. Bardasis, Phys. Rev. 159, 344 (1967).

[3] G. Lüders, Z. Physik 202, 8 (1967).

[4] B. Rosenblum and M. Cardona, Phys. Letters 13, 33 (1964).

[5] G. Fischer, R. Klein, and J. P. McEvoy, Solid State Commun. 4, 361 (1966).

[6] G. Fischer and R. Klein, Phys. Letters 23, 311 (1966).

[7] J. P. McEvoy, D. P. Jones, and J. G. Park, Solid State Commun. 5, 641 (1967).

[8] G. D. Cody and R. E. Miller, to be published.

[9] Y. Goldstein, in Proceedings of the Ninth International Conference on Low Temperature Physics, edited by J. G. Daunt, D. O. Edwards, F. J. Milford, and M. Yaqub (Plenum Press, New York, 1965), Pt. A, p. 400.

[10] D. Saint-James and P. G. de Gennes, Phys. Letters 7, 306 (1963).

[11] K. Maki, private communication.

[12] See in particular Fig. 1(a) of Ref. 5.

[13] K. Maki, Physics 1, 21 (1964), and Phys. Rev. 141, 331 (1966). The expressions derived in these papers are for $M(H)$ near H_{c2}, but similar formulas hold near H_{c3}.

[14] A. Rothwarf, J. I. Gittleman, and B. Rosenblum, Phys. Rev. 155, 370 (1967); see in particular Fig. 1. K. Maki, Phys. Rev. 141, 331 (1966); C. Caroli and K. Maki, Phys. Rev. 159, 306 (1967); K. Maki and G. Fischer, to be published.

[15] H. J. Fink and R. D. Kessinger, Phys. Rev. 140, A1937 (1965).

[16] A. Rothwarf, Phys. Letters 24A, 343 (1967). See also A. Houghton and F. B. McLean, Phys. Letters 19, 172 (1965).

[17] V. L. Ginzburg, Phys. Letters 13, 101 (1964).

[18] G. Fischer and R. Klein, to be published.

[19] We plan to publish shortly a more detailed investigation of this effect.

[20] A. A. Abrikosov, Zh. Eksperim. i Teor. Fiz. 47, 720 (1964) [translation: Soviet Phys.–JETP 20, 480 (1965)]; Maki and Fischer, Ref. 14.

[21] E. Helfand and N. R. Werthamer, Phys. Rev. 147, 288 (1966).

[22] N. R. Werthamer and W. L. McMillan, Phys. Rev. 158, 415 (1967).

[23] D. L. Decker, D. E. Mapother, and R. W. Shaw, Phys. Rev. 112, 1888 (1958).

PRECESSION MEASUREMENTS OF THE 0.847-MeV LEVEL OF Fe^{56} IMPLANTED IN IRON AFTER A (p,p') REACTION*

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A number of magnetic moments of excited states have been measured recently using the high internal hyperfine fields acting on the excited nuclei embedded in polarized iron. All levels measured until now have been reached either by radioactive decay,[1] by resonance fluorescence,[2] or by Coulomb excitation with backscattered heavy ions.[3] In this Letter we report the first results of an integral precession by an internal magnetic field of a level which was populated by a nuclear reaction.

The nuclear reaction mechanism not only enables one to reach levels which may not be accessible by the above mentioned methods, but, as demonstrated in the present work, no anomalies in the hyperfine fields are observed. Such anomalies are seen in the high recoil implantation work[4,5] and complicate the extraction of magnetic moments from the measured precession angles.

The experiment was carried out on the 0.847-MeV ($\tau = 10.6$ psec) first excited state of Fe^{56}. Since the g value of this state is well known,[1,2] measurements on it are an excellent test of possible anomalies appearing after implantation; in particular, the "conical-field"[5] and the "transient-field"[4] models may be tested. The excellent agreement of the present results with those of Refs. 1 and 2 indicate that in this case neither type of anomaly is present. The technique is therefore applicable to short-lived states, and reliable magnetic moments may be deduced from the measured precession angles.

The first excited 2^+ level of Fe^{56} was excited by inelastic protons, and the precession of this level in the internal field in polarized iron was measured. The excited iron nuclei obtained have an average recoil energy of about 0.25 MeV, which is nearly a hundred times lower than the recoil energy of about 23 MeV obtained by Coulomb excitation of the same level with backscattered 33-MeV oxygen ions. The value of the internal field of iron in iron obtained from the present "low-recoil" measurement is $H_{\text{int}} = (-0.43 \pm 0.14) \times 10^6$ G, in full agreement with the "static" value, $H_{\text{int}}(\text{Fe-Fe})$