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[6]Such field fluctuations exceed thermal fluctuations (calculated at the particle temperatures) by four or more orders of magnitude.

Apparent Universal Behavior of Fluctuation-Induced Diamagnetism in Superconductors*

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(Received 8 October 1970)

The dependence on temperature and magnetic field of the diamagnetism due to fluctuations above T_c, while showing marked deviations from the behavior expected on the basis of the Ginzburg-Landau theory, appears to follow a universal behavior if one introduces a new characteristic field H_s, which may be expressed in terms of other known material-dependent parameters.

We have previously reported[1] observation of a temperature-dependent diamagnetism above the critical temperature T_c due to thermal fluctuations of the superconducting order parameter Ψ. We have since extended these measurements to other materials (including type-II superconductors) and to much higher fields and temperatures; these results will be reported in detail elsewhere.[2] The purpose of this note is to show that these data are well described by an apparently universal function of appropriately scaled field and temperature variables. This empirically derived function deviates markedly from the behavior predicted[3,4] on the basis of the simple Ginzburg-Landau (GL) theory, especially for $T \gtrsim 2T_c$ and for fields comparable with $H_{c2}(0)$. These deviations demonstrate the expected breakdown of the GL theory in these regimes where short-wavelength fluctuations dominate, and where consequently the slow-variation approximations of the GL theory break down.[5] The apparent generality of our results suggests that there may exist a reasonably simple extension of the GL theory to deal with situations in which Ψ varies rapidly on the scale of $\xi(0)$, the zero-temperature GL coherence length. The attempt of Patton, Ambegaokar, and Wilkins[6] (PAW) to deal with the problem by means of an ad hoc cutoff energy is shown to have qualitative, but not quantitative, success.

Prange's calculation[4] of M', the magnetization due to thermal fluctuations, is exact within the framework of the GL theory. It is found that M' diverges at the temperature $T_{c2}(H)$, defined by the condition $H = H_{c2}(T_{c2})$, where the energy cost of a small fluctuation toward the superconducting state vanishes. Above T_{c2}, $M'(T)$ is predicted to fall off roughly as $(T-T_{c2})^{-1/2}$. In the temperature range near T_c where $H_{c2}(T)$ is linear in T, it is predicted that a field- and material-independent curve should be obtained if $M'/H^{1/2}T$ is plotted versus the dimensionless scaled temperature difference $(dH_{c2}/dT)(T-T_c)/H$. In particular, at the zero-field critical temperature T_c, it is predicted that

$$M'/H^{1/2}T_c = -0.323\Phi_0^{-3/2}k_B \qquad (1)$$

is a universal constant for all superconductors. Here Φ_0 is the flux quantum $hc/2e$. While our data show some of the qualitative features of Prange's result, they do not agree quantitatively nor do they scale so simply, except perhaps near T_{c2}.

The PAW theory attempts to correct for the overestimate in GL theory of short-wavelength fluctuations by introducing an unknown energy-cutoff parameter E into the fluctuation spectrum. As a result, they find that the magnetization is strongly depressed below the Prange value if $H > H^* \equiv mcE/\hbar e$ or if $T - T_c > T^* \equiv 4m\xi^2(0)T_c E/\hbar^2$, where $2m$ is the electronic pair mass. The expectation that the GL theory should break down when the characteristic wavelength over which the order parameter varies is smaller than $\xi(0)$ suggests that $E \approx \hbar^2/4m\xi^2(0)$, which corresponds to $H^* \approx H_{c2}(0)$ and $T^* \approx T_c$. Wavelengths as short as $\xi(0)$ occur for even the least energetic modes

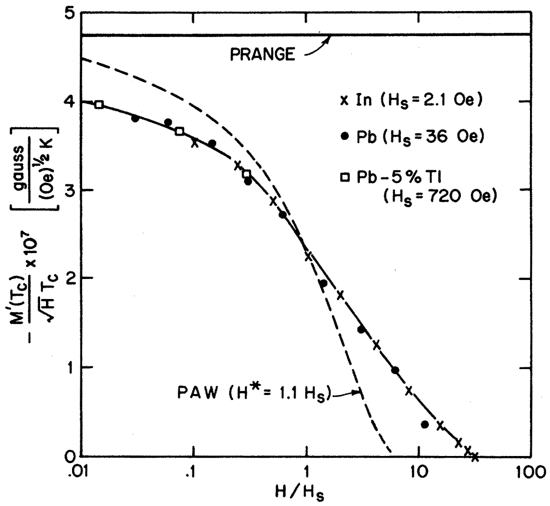

FIG. 1. The field dependence of $-M'(T_c)/H^{1/2}T_c$ plotted in units of the scaling field H_s and compared with the PAW falloff from Prange's theory.

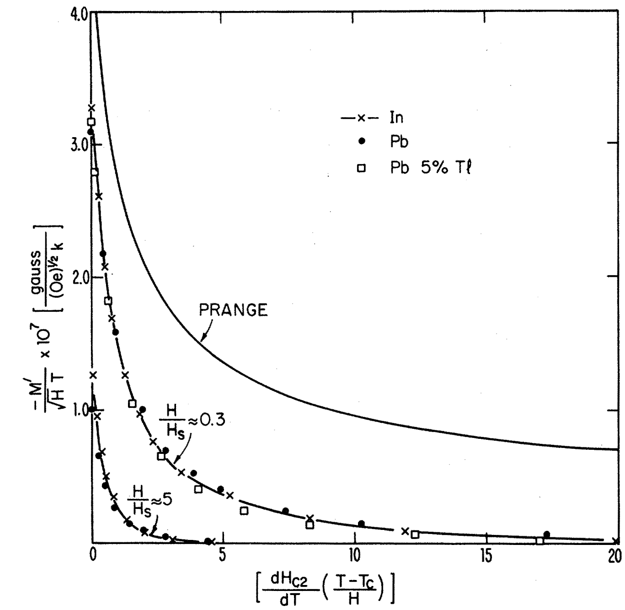

FIG. 2. Temperature dependence of the scaled magnetization of three materials at two values of H/H_s compared with the Prange result.

of fluctuations when $H \geq H_{c2}(0)$ but they occur at lower fields for the more energetic fluctuations, which vary more rapidly in space. Thus in the PAW calculation the cutoff does not become fully effective in reducing M' until the energies of even the lowest modes exceed the cutoff energy.

We have measured the temperature dependence of the magnetization at fields H up to 300 Oe and temperatures up to 16 K using instrumentation[2] based on a superconducting quantum interference magnetometer.[7] Our samples were long cylindrical single crystals (5×40 mm) of In, Pb, and PbTl alloys, grown from 99.9999% purity components. We take $M'(H, T) = M(H, T) - M(H, T \gg T_c)$ in order to eliminate the temperature-independent magnetization of the normal state. This value of M' is quite well defined since our present measurements extend far above T_c where M is constant within experimental error over a range of several degrees Kelvin except in the highest field measurements on Pb and PbTl, where a slightly temperature-dependent moment was still observable at 16 K. These more extensive measurements almost completely eliminate the baseline uncertainty which concealed the disagreement with the Prange theory in our earlier work.[1]

In Fig. 1 we present the field dependence of $M'(H, T_c)$ for three different materials. The ordinate $M'(T_c)/H^{1/2}T_c$ would be field independent if Prange's theory were valid. In fact the ordinate is progressively depressed as H increases. We find that for a given H, In deviates the most and Pb-5%Tl the least from Prange's theory. However, if the field is plotted in units of a scaling field H_s, which is chosen to be 2.1 Oe for In, 36 Oe for Pb, and 720 Oe for Pb-5%Tl, then the data for the three materials coincide and fall below Prange's theory in the same fashion. With our definition of H_s, the ordinate is half of Prange's value at $H/H_s = 1$; it is nearly zero when $H \approx 20H_s$, which incidentally is about equal to $H_{c2}(0)$ for the pure materials. The observed fall is very slow, being roughly logarithmic with field, so that the fall from 80 to 20% of the Prange value stretches over two decades in H/H_s. The PAW prediction is also shown, with H^* identified with $1.1H_s$ to force agreement at $H/H_s = 1$. It is not known whether the data would coincide with Prange's result in the limit as $H/H_s \to 0$, since accurate data at T_c can only be obtained in fields large enough to shift $T_c(H)$ down by at least the width of the transition due to sample inhomogeneity.

We also find that for a given value of H/H_s the temperature dependence of M' exhibits universal behavior. This is shown for two values of H/H_s in Fig. 2, where the temperature and magnetization are plotted in the scaled variables suggested by Prange's theory. The data lie substantially below Prange's theory but for a given H/H_s the temperature dependence is the same for all materials. We consider this universal behavior to be remarkable in view of the fact that the data involve clean type-I superconductors with both strong and relatively weak electron-phonon coupling and also a type-II alloy superconductor.

We conclude from these comparisons that to a good approximation, $M'(H, T)/H^{1/2}T$ depends only on two parameters, a scaled temperature $(dH_{c2}/dT)(T-T_c)/H$ and a scaled field H/H_s, where H_s is a single parameter for each material. The PAW theory suggests the correct sort of reduced variables to obtain universal behavior and qualitatively describes the suppression of fluctuations at high fields and high temperatures. However, in attempting to fit[8] the PAW theory to the data we are forced to use an $H^*(\approx H_s) \ll H_{c2}(0)$. With our interpretation of the cutoff energy E, this suggests an unreasonably long characteristic cutoff wavelength. An alternative, and perhaps more reasonable, viewpoint is that the PAW type of cutoff does not start to suppress the higher energy modes soon enough. Our data on the type-I superconductors show that for $H > H_{c2}(0)$, fluctuations have been almost completely suppressed, whereas, as noted before, the PAW cutoff [with $H^* \approx H_{c2}(0)$] only begins to suppress M' strongly when $H \approx H_{c2}(0)$.

The nature of the parameter H_s is not entirely understood. We find that $H_s/H_{c2}(0)$ is 0.05 for In, 0.06 for Pb, and 0.4 for Pb-5%Tl. Not only is H_s much less than $H_{c2}(0)$ for all three materials, but the alloy differs from pure lead more than a simple proportionality to $H_{c2}(0)$ would lead one to expect. Empirically, we find that to a good approximation

$$H_s = C(\Phi_0/2\pi\xi_0^2)(1+\xi_0/l)^2, \qquad (2)$$

where $C \approx 0.083$. This suggests that in accounting for the suppression of fluctuations by a magnetic field, the Pippard electrodynamic coherence length, defined by $1/\xi_P \approx 1/\xi_0 + 1/l$, may be the appropriate characteristic length, rather than $\xi_{GL}(0)$. The values of H_s for our samples, determined using (2) with ξ_0 and l determined by standard procedures,[9] are shown in Table I. Evidently the agreement is quite good, although it must be pointed out that the preference for ξ_P over $\xi_{GL}(0)$ depends solely on the data for one alloy. Moreover, as is clear from Fig. 1, the alloy data do not cover the lower part of the falloff because data at sufficiently high magnetic fields could not be conveniently obtained with our present apparatus. As a result, it is not established with certainty that the alloy data would follow the "universal" results for $H \gtrsim H_s$. For example, if M' for the alloy were also fully suppressed at $H_{c2}(0)$, it could not follow the universal behavior exhibited by the clean type-I superconductors. We are presently extending our measurements to other alloy systems and to higher values of H/H_s to check the dependence of H_s on l, and also to check whether the alloy samples follow the universal results even for $H > H_s$.

We feel that these results concerning the suppression of the diamagnetism at high fields contain considerable implicit information about the nature of the deviations from simple GL theory for short-wavelength phenomena. However, it is difficult to extract inductively from the data any quantitative modification of the GL free-energy functional or (equivalently) of the energy spectrum of the fluctuations, because the observed diamagnetism is the sum of contributions from many modes of excitation, each with its own spatial dependence. The best agreement

Table I. Superconducting material parameters for our samples.

Parameter	In	Pb	Pb-5%Tl	Unit
T_c	3.409[a]	7.2[a]	7.1[a]	K
$\kappa(T_c)$	0.0620[b]	0.328[c]	1.19[a]	
$(dH_{c2}/dT)_{T_c}$	13.4[d]	111[c]	402[a]	Oe/K
$H_{c2}(0)$	45[e]	594[c]	1800[f]	Oe
H_s (meas)	2.1 ± 0.2	36 ± 4	720 ± 150	Oe
H_s [from (2)]	2.07	36.4	616	Oe

[a] From this work.
[b] J. Feder and D. S. McLachlan, Phys. Rev. 177, 763 (1969).
[c] G. Fischer, Phys. Rev. Lett. 20, 268 (1968).
[d] $(dH_{c2}/dT)_{T_c} = \sqrt{2}\kappa(T_c)(dH_c/dT)_{T_c}$.
[e] Estimated using $\kappa(0) \approx 0.11$ (from Ref. b).
[f] Estimated from data on $H_{c2}(0)$ vs κ: G. Bon Mardion, B. B. Goodman, and A. Lacaze, J. Phys. Chem. Solids 26, 1143 (1965).

with GL occurs in the type-II alloy near T_{c2}; this is reasonable since as one approaches T_{c2}, the dominant contribution to the diamagnetism is from the very lowest modes, which are diverging there, and these lowest modes have the longest-wavelength spatial variation. On the other hand, far from T_{c2}, a great number of modes with much shorter wavelength make contributions comparable with the lowest ones, and the breakdown of the GL approximation is more severe. Because our sensitive measurements allow M' to be followed out to $2T_c$ and to high fields, and because they are obtained with rather ideal bulk samples, our data should allow quite a critical test for theoretical treatments which go beyond the region near the critical point.

*Work supported in part by the National Science Foundation, the U. S. Office of Naval Research, the Advanced Research Projects Agency, and the Joint Services Electronics Program.

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[8]It is interesting to note that the PAW curve agrees quite well with the data if the horizontal scale is expanded by a factor of 2; i.e., if (H/H_s) were replaced by $(H/H_s)^{1/2}$ in their results. Although this observation may well give guidance on how to improve their model, no rationale for such a change is presently available.

[9]For the pure materials, where $l \gg \xi_0$, H_s depends only on ξ_0. This parameter was evaluated using the standards results $\xi(T) = 0.74\xi_0\,(1-T/T_c)^{-1/2}$ and $H_{c2}(t) = \Phi_0/2\pi\xi^2(T)$. These give the relation $T_c dH_{c2}/dT|_{T_c} = \Phi_0/2\pi(0.74)^2\xi_0^2$, from which ξ_0 can be determined using the known values of dH_{c2}/dT and T_c. For the alloy, H_s is calculated by multiplying the value of H_s computed for pure lead by the factor $(1+\xi_0/l)^2$. The required ratio of ξ_0/l can be determined from the relation $(T_c dH_{c2}/dT)_{\rm Pb}/(T_c dH_{c2}/dT)_{\rm PbTl} = \chi(x)$, where $\chi(x)$ is a known function [see Ref. 5, p. 338] of ξ_0/l. These procedures yield $\xi_0(\mathrm{In}) = 3640$ Å, $\xi_0(\mathrm{Pb}) = 870$ Å, and $\xi_0/l = 3.12$ for the alloy, and the values of H_s shown in Table I.

Hypersound Attenuation in Superconductors by Quasiparticle Creation*

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The attenuation of 10-GHz longitudinal sound waves in superconducting molybdenum ($T_c = 0.914$ K) and cadmium ($T_c = 0.500$ K) shows the high-frequency behavior predicted by the BCS theory. In particular, the onset of the rapid drop in attenuation with decreasing temperature that is characteristic of superconductors is shifted downward to the temperature T_ν (0.905 K in molybdenum and 0.490 K in cadmium) at which the superconducting energy gap equals the phonon energy. The analysis of the measurements indicates a large anisotropy in the energy gaps of both metals.

In the original publication of the BCS theory of superconductivity,[1] the low-frequency limit (i.e., phonon energy $h\nu$ small compared with the energy gap 2Δ) for the attenuation of sound was written as

$$\alpha_S/\alpha_N = 2f(\Delta), \tag{1}$$

where α_S and α_N are the acoustic attenuations in the superconducting and normal states, and f is the Fermi function. For higher frequencies the results include contributions to the phonon absorption not