MAGNETIC PROPERTIES OF SUPERCONDUCTING THIN FILMS IN THE NONLOCAL REGIME*

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One well-known prediction of the microscopic theory of superconductivity is a nonlocal current-field relationship, viz., $j(x) = -(c/4\pi) \times \int K(x,x')A(x')dx'$ in the weak-field limit. In a thin film of thickness $d \lesssim \xi$ placed in a magnetic field $\vec{H}$ parallel to its surface, the vector potential A is constrained to vary appreciably over the range of ξ of the kernel K, so that nonlocality must be considered. In a pure material $\xi_0 = 0.18\, v_F/T_c$; in an impure sample ξ is reduced to $[\xi_0^{-1} + l^{-1}]^{-1}$, where $l = v_F \tau_{tr}$ is the mean free path for bulk scattering. Strong field effects have previously been calculated in the local limit in two situations: (a) Near the transition temperature T_c the Ginzburg-Landau-Gor'kov[1] (GLG) theory determines the critical field, and (b) in "very dirty" superconducting films Maki[2] has found the energy spectrum for all temperatures and fields. Using the same thermodynamic Green's function techniques[3] and assuming the order parameter Δ to be constant,[4] we have extended these results to the nonlocal regime, thus encompassing common experimental situations characterized by significant deviations from the thickness dependence predicted by the local theories. Previous analyses of relevant data[5,6] near T_c had no firm microscopic basis. In case (b) the restrictions $l \ll d \ll (\xi_0 l)^{1/2}$ precluded any meaningful comparison of the original theory with experiment. These obstacles have now been removed.

The Gor'kov integral equations[3] are iterated with respect to the perturbation $X = (e/mc)\vec{A}\cdot\vec{p}$. The conditions for the rapid convergence of this expansion, obtained by evaluating the ratio of successive terms at the critical field H_c, are stated in the caption of Fig. 1. The boundaries of the film are at $x = 0$ and $x = d$. With $\vec{H}$ applied along the z axis, $\vec{j}$ and $\vec{A}$ have y components only and are antisymmetric about $x = d/2$ in the gauge where Δ is real.

The Green's functions $G_\nu(\vec{r},\vec{r}')$ and $F_\nu(\vec{r},\vec{r}')$ are expressed in terms of the (real) normal-state eigenfunctions w_n of the sample in zero field. Sums over intermediate states can be performed by means of a trick used by De Gennes[7] in a similar connection. Thus the A^2 contribution to the right-hand side of the linearized self-consistency condition $\Delta = gT\sum_\nu F_\nu$ (averaged over the volume V of the sample),

$$F_\nu^{(2)} = \Delta V^{-1}\int d^3r \iiint d^3s\, d^3u\, d^3v [G_{-\nu}^{(0)}(\vec{s},\vec{r}) \times G_\nu^{(0)}(\vec{s},\vec{u})X(\vec{u})G_\nu^{(0)}(\vec{u},\vec{v})X(\vec{v})G_\nu^{(0)}(\vec{v},\vec{r}) + 2 \text{ similar terms}],$$

can be transformed as follows [$\omega_\nu = (2\nu+1)\pi T$]:

$$F_\nu^{(2)} = \Delta V^{-1}\sum_{mn} \frac{\langle m|X|n\rangle\langle n|X|m\rangle}{(\epsilon_n + i\omega_\nu)(\epsilon_n - i\omega_\nu)^2(\epsilon_m - i\omega_\nu)} + \cdots$$

$$= -\Delta \frac{\pi N(0)}{\omega_\nu^2}\int_{-\infty}^{\infty} dt \exp(-2|\omega_\nu t|)\langle X(0)X(t)\rangle.$$

The average $\langle X(0)X(t)\rangle$ is evaluated over all classical trajectories of electrons at the Fermi surface linking two points $\vec{r},\vec{r}'$ inside the sample in time t. The final result may be written as

$$-gN(0)\Delta\tfrac{1}{3}\left(\frac{ev_F}{c}\right)^2 d^{-1} \times \sum_\nu \int_0^d dx \int dx' A(x)K_\nu(x-x')A(x').$$

Similar manipulations give

$$j(x) = -\tfrac{2}{3}(ev_F\Delta)^2 N(0) c^{-1}\sum_\nu \int dx' K_\nu(x-x')A(x').$$

The integration over x' extends from 0 to d if the boundary scattering is diffuse and from $-\infty$ to ∞ if electrons are specualrly reflected. The results below correspond to the latter, simpler, case. $A(x)$ is then defined throughout space by its Fourier cosine series.

Near T_c this approach justifies Bardeen's nonlocal generalization of the GL equations.[8]

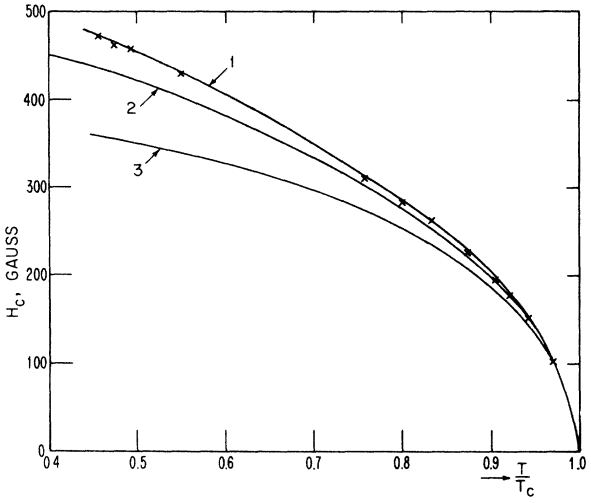

FIG. 1. Temperature dependence of the critical field of a superconducting alloy film. Crosses: Toxen's data for a 2220Å-thick In-4.6 at.% Sn film. Theoretical curves: (1) Formula (8) including terms in H^4, valid when $1-T/T_c \ll \xi_0/\xi$ and $d\xi_0/\xi^2$. (2) Generalized Maki formula (7), valid when $l \ll \xi_0$ and $(d\xi_0)^{1/2}$. (3) Generalized Gor'kov formula (5), valid when $1-T/T_c \ll 1$ and $d\xi_0/\xi^2$. All curves were adjusted to fit near T_c.

The field distribution is determined by

$$\left(\frac{4\pi}{c}\right)j(k_m) = -\left(\frac{\Delta^2}{\lambda_L^2}\right)\sum_{\nu=0}^{\infty} K_\nu(k_m)A(k_m) \equiv -\psi^2 K(k_m)A(k_m), \tag{1}$$

while the reduced order parameter $\psi = \Delta(H,T)/\Delta(0,T)$ satisfies

$$\psi^2 = 1 - \sum_{m=0}^{\infty}\left(\frac{A(k_m)}{2H_{cb}(T)}\right)^2 K(k_m). \tag{2}$$

Here

$$A(k_m) = \left(\frac{2}{d}\right)\int_0^d A(x)\cos(k_m x)dx,$$

$k_m = (2m+1)\pi/d$, $H_{cb}(T)$ is the bulk thermodynamic critical field,

$$K_\nu(k) = 2\pi T\frac{f(k\xi_\nu)}{\omega_\nu^2}\left\{\omega_\nu + \tfrac{1}{2}\left[\frac{1}{\tau} - \frac{f(k\xi_\nu)}{\tau_1}\right]\right\}^{-1}, \tag{3}$$

$$\xi_\nu = \frac{v_F}{2\omega_\nu + \tau^{-1}},$$

$\tau_{tr}^{-1} = \tau^{-1} - \tau_1^{-1}$ is the inverse transport relaxation time while τ is the single-electron relaxation time. The function

$$f(z) = \tfrac{3}{2}\int_0^1 \frac{1-\mu^2}{1+(z\mu)^2}d\mu = \tfrac{3}{2}z^{-1}[(1+z^{-2})\tan^{-1}z - z^{-1}] \tag{4}$$

arises because of angular averaging; it also appears in the Pippard kernel,[8] viz., $K_p(k) = (\xi/\xi_0)[f(k\xi)/\lambda_L^2(T)]$. $K(k)$ as defined in (1) is the asymptotic form of the weak-field bulk kernel for $\Delta \to 0$ [compare with Eq. (37.15) in reference 3]. The vertex corrections appearing in (3) have been evaluated exactly in this limit. In practice, however, only τ_{tr} can be determined; if one assumes $\tau = \tau_{tr}$, K reduces to a sum of Pippard kernels, provided the substitutions $\xi \to \xi_\nu \equiv [(2\nu+1)\xi_T^{-1} + l^{-1}]^{-1}$ and $\xi_0 \to \xi_T \equiv v_F/2\pi T = 0.88(T_c/T)\xi_0$ are made.

The critical field H_c for a second-order transition is deduced by setting $\Delta = 0$ in (2) and taking $A(z) = H_c(x-d/2)$, $A(k_m) \propto (2m+1)^{-2}$. Since the summation over m in (2) differs from its first term by no more than 2%, a convenient expression follows if $m > 0$ terms are neglected:

$$H_c = \frac{\pi^2}{8}\frac{c}{ed}\left(3\ln\frac{T_c}{T}\right)^{1/2}\left[\sum_{\nu=0}^{\infty}\frac{\xi_T\xi_\nu}{(2\nu+1)^2}f\left(\frac{\pi\xi_\nu}{d}\right)\right]^{-1/2} \equiv \frac{\pi^2}{2}\frac{\lambda(d)}{d}H_{cb}. \tag{5}$$

$\lambda(d) = [K(\pi/d)]^{-1/2}$ can be interpreted as an effective penetration depth containing nonlocality effects. Toxen[5] assumed that nonlinear effects could be treated separately and used an alternative definition of $\lambda(d)$ which give the correct weak-field susceptibility. For a given kernel, both results would agree within 2% for $d \lesssim d_c$. However, a computer is not required to determine $\lambda(d)$ in our case. The critical thickness d_c above which the thermodynamic transition changes from second to first order is easily obtained from (1) and (2), viz., $d_c = (\pi/\sqrt{2})\lambda(d_c)$ and, hence, $H_c(d_c)/H_{cb} = 2.20$ in agreement with Baldwin's experiments on the onset of hysteresis in tin films.[6]

Our extension of Maki's dirty-film theory[2] to the nonlocal regime proceeds along similar lines. Assuming s-wave scattering, we perform the impurity averaging at the outset and obtain equations identical in form to those for a pure superconductor. The Green's functions are conveniently expressed in terms of the w_n's

of the pure film if the spatial dependence of $F_\nu(x,x)$ and $G_\nu(x,x)$ is negligible.[9] The net result of our calculations is that Maki's formulas retain their validity outside the local limit $l \ll d$ provided his depairing parameter $\zeta = 2\tau\alpha/\Delta$ is redefined as follows:

$$\alpha = \tfrac{1}{6}\sum_m \left(\frac{ev_F A(k_m)}{c}\right)^2 f(k_m l). \qquad (6)$$

In particular H_c is given by (neglecting $m > 0$ terms as before)

$$\ln\frac{T_c}{T} = \psi\left[\tfrac{1}{2} + \frac{2\tau}{3\pi T}\left(\frac{2eH_c v_F d}{\pi^2 c}\right)^2 f\left(\frac{\pi l}{d}\right)\right] - \psi(\tfrac{1}{2}), \qquad (7)$$

where ψ is the di-gamma function. The tunneling density of states $\rho(\omega)$ is the same as calculated by Skalski, Betbeder-Matibet, and Weiss[10] in a different physical context with their Γ set equal to $2\tau\alpha$. The energy gap ω_g disappears for $\zeta > 1$. Unfortunately Douglass and Meservey's measurements on Al-Pb junctions[11] cannot be readily compared with theory. Their operational determination of the "energy gap" of aluminum from the I-V characteristic actually locates the peak in $\rho(\omega)(e^{\omega/T}+1)^{-1}$ and systematically overestimates ω_g for increasing H. The magnetic field dependence of the dynamic conductance dI/dV of thin film sandwiches would be much easier to interpret. Since the restrictions $(l/\xi_0)l \ll d \ll (\xi_0 l)^{1/2}$ could be met reasonably well in high-ξ_0 materials, experimental tests of the extended theory should be possible.

The limiting expressions for H_c obtained by De Gennes and Tinkham and Shapoval[7] agree with ours whenever $(eH_c d\xi/c)^2 \ll 1$. The corresponding restrictions are somewhat different from theirs; near T_c, in particular, we find the experimentally observed behavior[5,6] $H_c \propto (1-T/T_c)^{1/2} d^{-3/2}$ for $d^2 \ll \xi^2 \ll d\xi_0(1-T/T_c)^{-1}$.

If $l \sim \xi_0$, considerable improvement over (5) and (7) can be achieved at intermediate temperatures by including terms in H^4. Such a calculation gives[12]

$$\ln\frac{T_c}{T} = 2\sum_{\nu=0}^{\infty} \frac{1}{(2\nu+1)\xi_\nu} \frac{\gamma}{(2\nu+1)\xi_T^{-1} + \gamma l^{-1}}, \qquad (8)$$

where $\gamma = \beta\tfrac{2}{3} f(\pi\xi_\nu/d) - \beta^2 \tfrac{2}{5}[4f'(2\pi\xi_\nu/d) - f'(\pi\xi_\nu/d)]$; $\beta = (4eH_c d\xi_\nu/\pi^2 c)^2$ is the natural expansion parameter while $f'(z) = (5/4)\{f(z) + z^{-2}[f(z)-1]\}$ has the same asymptotic properties as $f(z)$.

A comparison with Toxen's data on a 2220Å-thick In-4.6 at.% Sn films is shown in Fig. 1. ξ_0 was first determined from a fit to (5) near T_c, viz., 2000 Å taking $l = 840$ Å, corresponding to $\rho l = 1.2\times10^{-11}$ Ω cm^2. The same procedure gives $\xi_0 = 2250$ Å for pure In films. The decreased value of ξ_0 found in the alloy agrees with that expected from the observed 12% increase in T_c and the 1.5% increase in v_F given by the free-electron model. Such effects were not consistently taken into account by Toxen. A more realistic fit based on the analog of (5) for diffuse scattering gives $\xi_0 = 2700$ Å and $\rho l = 1.1\times10^{-11}$ Ω cm^2 for pure In.

Calculational details, additional applications, as well as an extensive analysis of experimental data will be published elsewhere. The authors would like to thank Professor P. C. Martin for many helpful discussions and Dr. A. M. Toxen for kindly forwarding us some of his data.

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[9]This assumption is implicit in Maki's derivation. It is valid at all T provided $d \ll (\xi_0 l)^{1/2}$ or $H_c \gg H_{c2}$, as can be surmised from the work of P. G. de Gennes, reference 7, or E. Helfand and N. R. Werthamer, Phys. Rev. Letters 13, 686 (1964). A rival local theory proposed by G. Rickayzen, Phys. Rev. 138, A73 (1965), assumes Δ constant, but allows $F_\nu(x,x)$ to vary. One easily verifies that his final solution implies that $\Delta(x) \propto \sum F_\nu(x,x)$ does vary appreciably across the

film (except near T_c, where the results coincide with Maki's) in contradiction with the original assumption.

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RELATIVISTIC CORRECTIONS TO THE TRITON BINDING ENERGY

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We present here the results on the relativistic corrections to the triton binding energy, using the three-body techniques developed by this group in recent years. To our knowledge, no reliable estimate of the relativistic effects exists so far, though general considerations suggest that these corrections could contribute anything between 10 and 20% to the triton energy.[1] It has also been argued that the relativistic corrections could be of the order of hard-core effects.[2] Perhaps the lack of a precise estimate should be attributed to the absence of an accurate triton wave function which, though a prerequisite for the calculation of small effects, could not have been provided by the variational techniques usually adopted for the calculation of triton parameters. On the other hand, the present method, which uses separable two-body potentials,[3] leads to an exact expression for the three-body wave function, and is therefore ideally suited for the calculation of relativistic and similar effects in a perturbative manner. Our results not only confirm that a perturbative procedure is justified, but that the magnitude of the total relativistic correction is small, being about $\frac{1}{2}$ MeV.

The main question, of course, hinges on the choice of a good zeroth-order wave function which can be made the basis of a perturbative calculation. Now insofar as our techniques, as well as similar ones developed by other authors,[4,5] reduce the solution of a three-body problem to that of a set of coupled one-dimensional Fredholm equations, the evaluation of an accurate wave function is limited only by practical considerations of a manageable number of such equations. With pure s-wave interactions, this number is two, but neither are the two-body data well represented, nor is the three-body wave function accurately determined (as it gives too high a binding energy).[6] The least that is required to give a fairly detailed fit to two-body data is a potential with tensor forces. With a Yamaguchi type potential,[7] which has two form factors for the triplet interaction (associated with s and d states, respectively) and one for the singlet interaction (s state), the three-body wave function depends on three one-dimensional "spectator functions."[8]

Using such a wave function, Bhakar and Mitra[9] recently obtained the triton binding energy and the percentage probabilities of various spectroscopic states for the cases of Yamaguchi[7] and Naqvi[10,11] separable potentials. The results clearly brought out the decisive role of the tensor force in effecting a much needed reduction in the triton binding energy over the corresponding figure for purely s-wave interaction. The best results, which were obtained with the pure Naqvi set,[10,11] put the H^3 binding energy at 8.85 MeV, a clear 1-MeV lead (in terms of proximity to the experimental value of 8.48 MeV) over the next-best figure of 9.95 MeV obtained with a combination of Yamaguchi-triplet[7] and Naqvi-singlet[10] potentials. Now the complete Naqvi potential, in addition to a Yamaguchi-type structure,[7] has a spin-orbit force in the triplet d state[11] and another separable term in the singlet d state,[10] which are necessary for giving a detailed fit to the deuteron and N-N scattering data. To the extent that the effect of these two terms on the binding energy had not been estimated in reference 9, it was not then possible to assert that the pure Naqvi set of parameters was indeed superior