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Fluctuation-Induced Diamagnetism in Dirty Three-Dimensional, Two-Dimensional, and Layered Superconductors*

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The fluctuation-induced diamagnetism in dirty layered superconductors is calculated for the case in which the magnetic field is perpendicular to the layers. The tight-binding model of Lawrence and Doniach and the microscopic theory of Maki and Takayama are employed. A calculation is also performed for dirty two-dimensional thin films, and a parameter is identified that interpolates between the two- and three-dimensional behavior for layered superconductors.

I. INTRODUCTION

Experimental investigations by Gamble, Geballe, DiSalvo, and Klemm[1a] and by Geballe *et al.*,[1b] have led to a class of layered superconductors suitable for both experimental and theoretical study. These materials were first investigated theoretically by Lawrence and Doniach,[2] and later by Tsuzuki,[3] who used the tight-binding model within the framework of Schmid[4] to calculate the conductivity and susceptibility due to fluctuations above T_c in these quasi two-dimensional materials. Their theory was a simple Ginzburg-Landau type, and took no account of the time dependence of the fluctuations. It predicted that the layered superconductors should have a temperature dependence in zero magnetic field which behaved three dimensionally just above T_c, and two dimensionally further from T_c. Although the experimental results obtained to this point are not definitive, reliable experimental results should soon be available. For comparison with the experiments, more reliable theoretical calculations are desirable.

In this paper we report calculations based on the microscopic framework of Maki and Takayama[5] for these layered materials when the magnetic field is directed perpendicular to the layers. Since the materials of interest, transition-metal dichalcogenide intercalates, are formed by the intercalation of organic molecules between the layers of the transition-metal dichalcogenides, the layers exhibit various structural and chemical defects, and therefore we expect the materials to be well within the dirty limit. In fact, preliminary experimental investigations seem to indicate that these materials are extremely dirty,[6,7] so much so that we must be careful that the mean free paths are not so short that the whole formalism breaks down. Fortunately, the $E_F\tau$ deduced from the measurements seems large enough for the theory to be valid. For comparison, we will also perform the calculation for two-dimensional thin films. We further note that the calculation has recently been carried out by Gerhardts and Doniach[8] in the clean limit, where nonlocal effects are important.

The so-called "zero-point" term will have to be investigated both for the two-dimensional case and for the layered superconductors, because it is not obvious that the reasons for dropping the three-dimensional analog also apply to these two cases. Maki and Takayama[5] (MT) dropped this term, and found that the theory predicted a field dependence of the magnetization in good agreement with experiment, whereas the theories of Lee and Payne[9] and Kurkijärvi, Ambegaokar, and Eilenberger,[10] which were the same as MT in all respects except for the inclusion of this term, were not quantitatively in agreement with experiment. Maki[11] has demonstrated that this term is weakly dependent upon both field and temperature and therefore not detected in the present experiments. We will show that the zero-point terms are small in the two-dimensional and layered cases as well.

To avoid unnecessary mathematical approximations, we will evaluate the magnetization using the full dirty-limit propagator, instead of the Ginzburg-Landau approximation for it. We shall see that the agreement of the correct expression for the temperature dependence is good. We note that, since the temperature dependence of the magnetization depends upon the dimensionality of the system, it would be helpful for understanding layered superconductors to have a theory that predicts the temperature dependence reliably.

Since for the layered and two-dimensional cases the magnetic field is directed perpendicularly to the conducting plane(s), the formalism for the dirty limit based upon the formation of Landau orbits perpendicular to the magnetic field is still valid. The Cooper pairs may complete any number of Landau orbits without having to leave the plane in which they began. Motion in the direction of the field is described by a function of the single variable k_z, since the effective mass is assumed to be diagonal in the basis of the states directed parallel and perpendicular to the planes. The

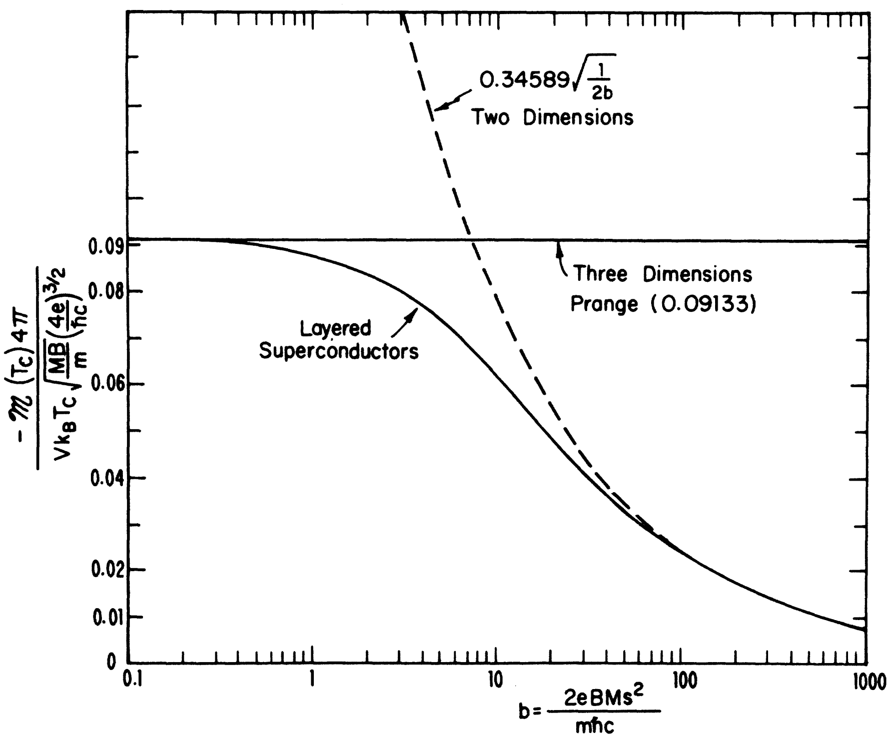

FIG. 1. Plot of static three-dimensional (Prange), two-dimensional, and layered magnetizations as a function of $b=\sigma^{-1}$. For the two-dimensional case, d is replaced by s for comparison. [See Eqs. (24)–(26).]

thickness d of a two-dimensional thin film is much less than the coherence length $\xi(0)$. Thus, we may use only the lowest k_z value allowed. For the layered superconductors, we use the tight-binding form of Lawrence and Doniach,[2] based upon Josephson coupling between the layers. For formal purposes, we define a quantity $e(k_z)$ such that

$$\begin{aligned} e(k_z) &= (m/M)\,k_z^2 && \text{anisotropic three-dimensional superconductors} \\ &= U[1-\cos(k_z s)] && \text{layered superconductors} \\ &= 0 && \text{two-dimensional thin films,} \end{aligned} \tag{1}$$

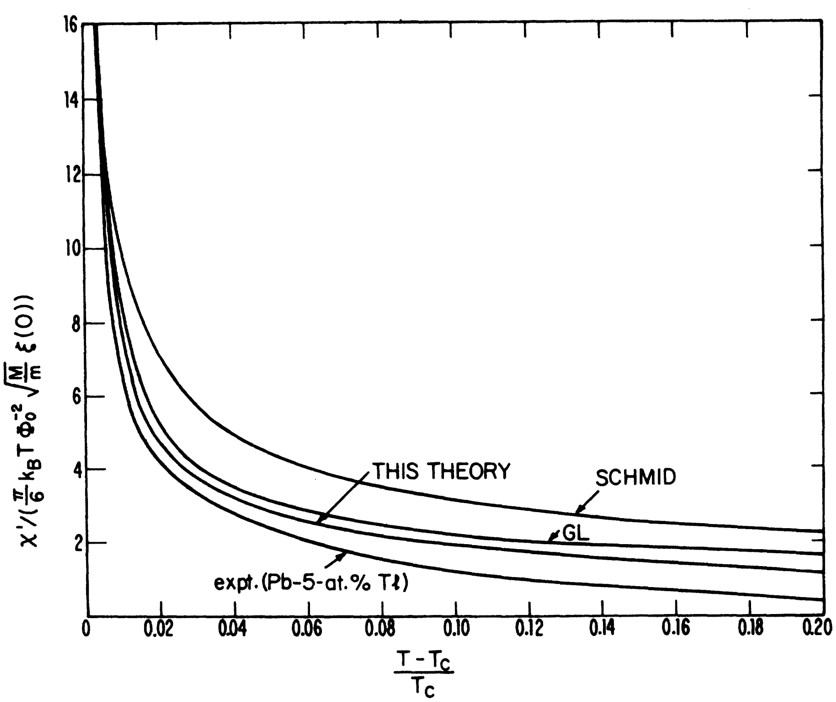

FIG. 2. Zero-field susceptibility for three-dimensional alloys from Eq. (20) plotted along with the Schmid theory, the Ginzburg-Landau approximation to Eq. (20), and the extrapolated experimental curve for Pb-5-at.% Tl alloys (Ref. 13).

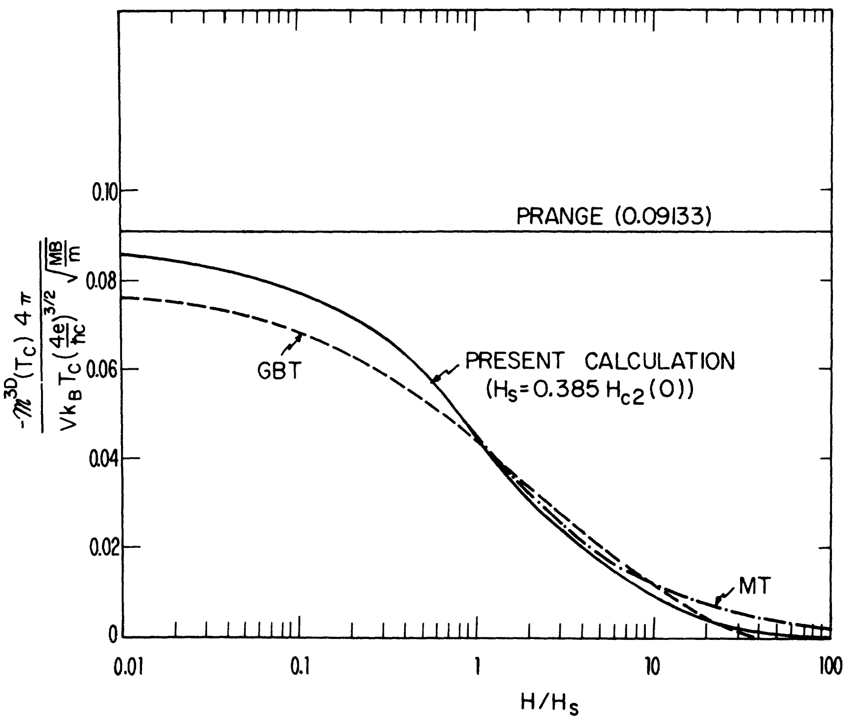

FIG. 3. Comparison of the full propagator theory with the experiment of GBT (Ref. 13) for Pb-5-at.% Tl and the approximate calculation of Maki and Takayama (Ref. 5) for the magnetic field dependence of three-dimensional superconductors at T_{c0}.

where U is defined such that as $k_z \to 0$, $e(k_z)$ for the layered materials approaches $(m/M)k_z^2$; $U \equiv 2m/(Ms^2)$. We note that for three dimensions, k_z varies from $-\infty$ to $+\infty$; for two dimensions, it is between $\pm\pi/d$; and for the layered materials, it varies between $\pm\pi/s$. M and m are the effective masses parallel and perpendicular to the magnetic field, respectively; d is the thickness of the thin film; and s is the spatial period of a layered material. In our calculations, we shall use the natural units of $c=\hbar=k_B=1$, restoring these quantities in the final equations.

II. THEORY

The free energy due to fluctuations in the presence of magnetic field H is obtained by a coupling-constant integration of the approximate potential energy

$$\frac{F}{V}=-\frac{TeH}{\pi}\sum_{\omega_\nu=-\infty}^{\infty} e^{i\omega_\nu 0^+} \times\sum_{n=0}^{\infty}\int\frac{dk_z}{2\pi}\ln\{\mathfrak{D}(n,k_z,\omega_\nu)\} \; . \tag{2}$$

Here $\mathfrak{D}(n,k_z,\omega)$, the fluctuation propagator in the dirty limit, is equivalent to the sum of ladder diagrams

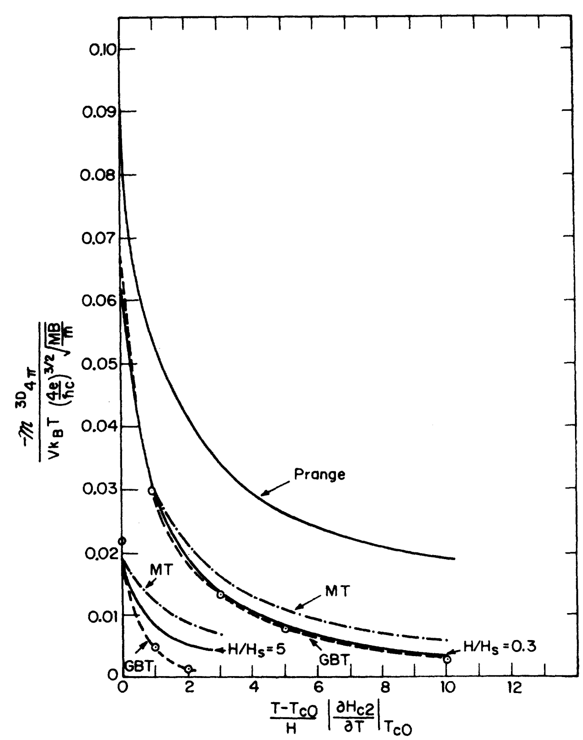

FIG. 4. Temperature dependence of three-dimensional superconductors. Plotted are the full propagator calculations, the approximate calculation of Maki and Takayama (Ref. 5), and the experimental data of GBT (Ref. 13) for Pb-5-at.% Tl.

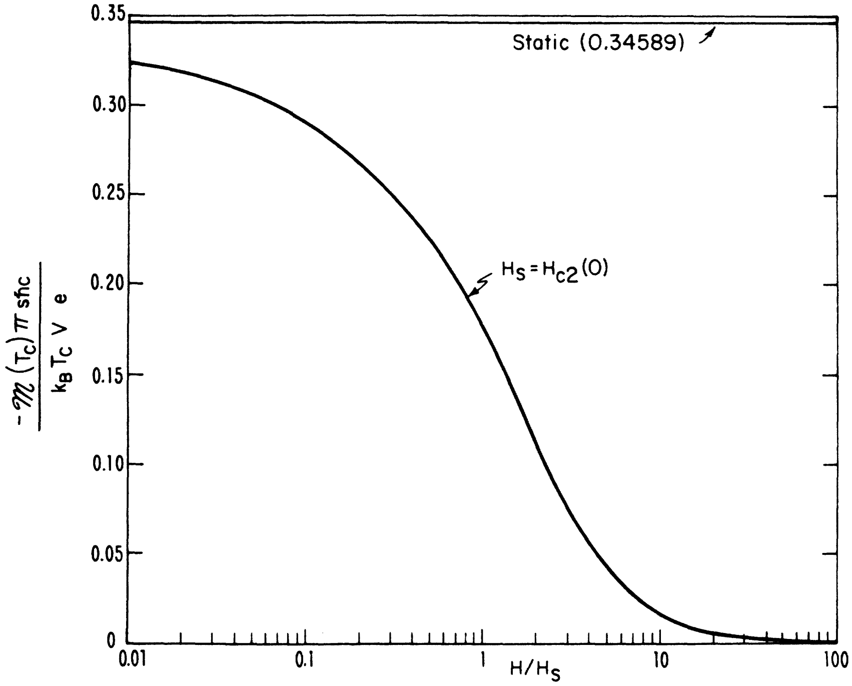

FIG. 5. Field dependence of two-dimensional thin films. Plotted are the static value and the full propagator theory at T_{c0}. Note that the static value is independent of the magnetic field.

$$\mathfrak{D}^{-1}(n,k_z,\omega_\nu)=|g|N(0)\times\left[\psi\left(\frac{1}{2}+\frac{|\omega_\nu|+D[4eH(n+\frac{1}{2})+e(k_z)]}{4\pi T}\right)-\psi\left(\frac{1}{2}-\frac{\epsilon_0}{4\pi T}\right)\right]\;, \tag{3}$$

where $N(0)$ is the electron-state density for a single spin direction, $D=\frac{1}{2}lv$ is the two-dimensional diffusion constant perpendicular to the magnetic field, g is the coupling constant, $\psi(z)$ is the digamma function, and ϵ_0 is determined by

$$-\ln(T/T_{c0})=\psi(\tfrac{1}{2}-\epsilon_0/4\pi T)-\psi(\tfrac{1}{2})\;, \tag{4}$$

where T_{c0} is the zero-field transition temperature and ω_ν is the Matsubara frequency defined by

$$\omega_\nu=2\pi\nu T\;. \tag{5}$$

Equation (3) can be derived from the microscopic theory when the electron scatters many times in a given layer before tunneling to an adjacent layer.

By analytic continuation, we may transform the above sum over the Matsubara frequency ω_ν into an integral over ω:

$$\frac{F}{V}=\frac{-eH}{\pi^3}\sum_{n=0}^{\infty}\int dk_z\int_0^{\infty}d\omega\left(\frac{1}{2}+\frac{1}{e^{\beta\omega}-1}\right)\times\mathrm{Im}[\ln\mathfrak{D}^{-1}(n,k_z,i\omega)]\;, \tag{6}$$

where $\beta=1/T$ is the inverse temperature. We define a function f such that

$$f(x)=\ln\{|g|N(0)[\psi(\tfrac{1}{2}+x)-\psi(\tfrac{1}{2}-\epsilon_0/4\pi T)]\} \tag{7}$$

and $f^{(n)}(x)$ is the nth derivative of $f(x)$. We then use the Poisson sum formula, differentiate with respect to H to obtain the magnetization, integrate by parts three times with respect to the dummy variable x (following Prange[12] and MT[5]), obtaining

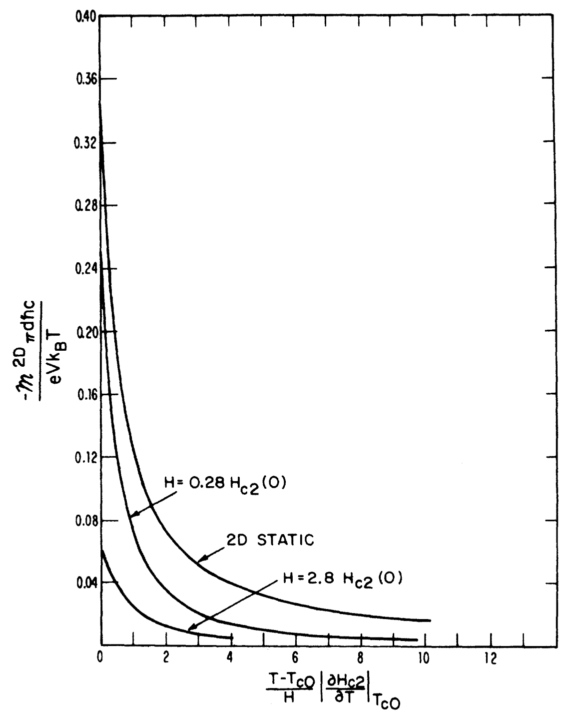

FIG. 6. Temperature dependence of two-dimensional thin films for $H=0.28H_{c2}(0)$ and $H=2.8H_{c2}(0)$. Plotted also is the static function ("two-dimensional Prange").

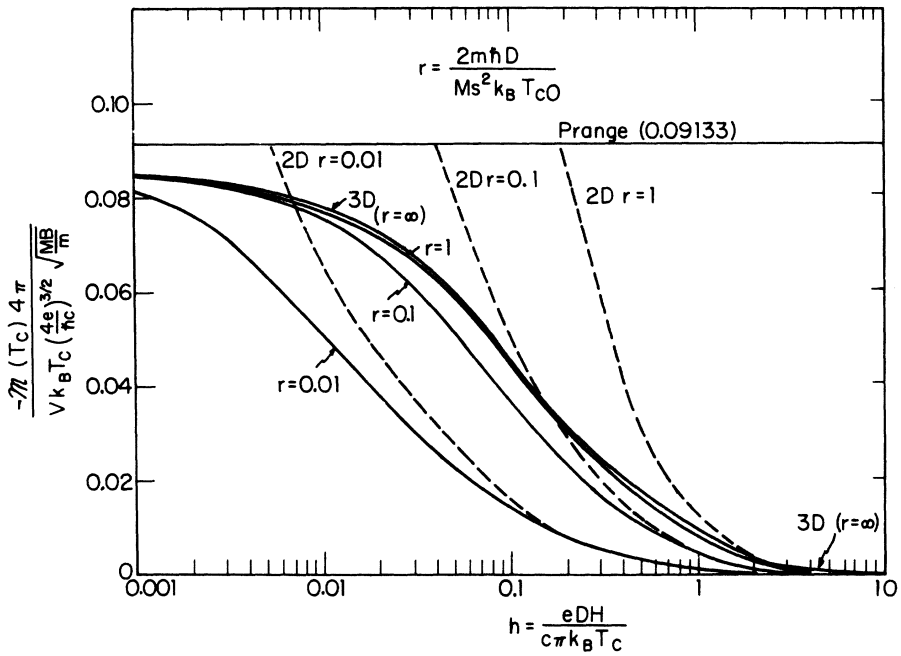

FIG. 7. Comparison of the field dependence for layered superconductors at T_{c0} with the two- and three-dimensional field dependences at T_{c0}. Plotted is the layered magnetization for $r=1$, 0.1, and 0.01, the Prange value, the three-dimensional curve from Fig. 3, and the high-field parts of the two-dimensional field dependences (from Fig. 5) multiplied by $(\frac{1}{2}\sigma)^{1/2}$ for comparison.

$$\mathfrak{M} = \frac{2eV}{\pi^2}\int\frac{dk_z}{2\pi}\int_0^\infty d\omega\left(\frac{1}{2}+\frac{1}{e^{\beta\omega}-1}\right)$$
$$\times\,\mathrm{Im}\left(\frac{h}{12}f^{(1)}\left(\frac{i\omega}{4\pi T}+\frac{h}{2}+\frac{De(k_z)}{4\pi T}\right)-\frac{h^2}{24}f^{(2)}\left(\frac{i\omega}{4\pi T}+\frac{h}{2}+\frac{De(k_z)}{4\pi T}\right)\right.$$
$$\left.-\frac{1}{2}\sum_{n=1}^{\infty}\int_{-1/2}^{1/2}dx\,(x^2-\tfrac{1}{12})\left\{\frac{d^3}{dy^3}\left[yf\left(\frac{i\omega}{4\pi T}+hy+\frac{De(k_z)}{4\pi T}\right)\right]\right\}\right), \qquad (8)$$

where $y=x+n$ is taken after differentiation, and h is defined by

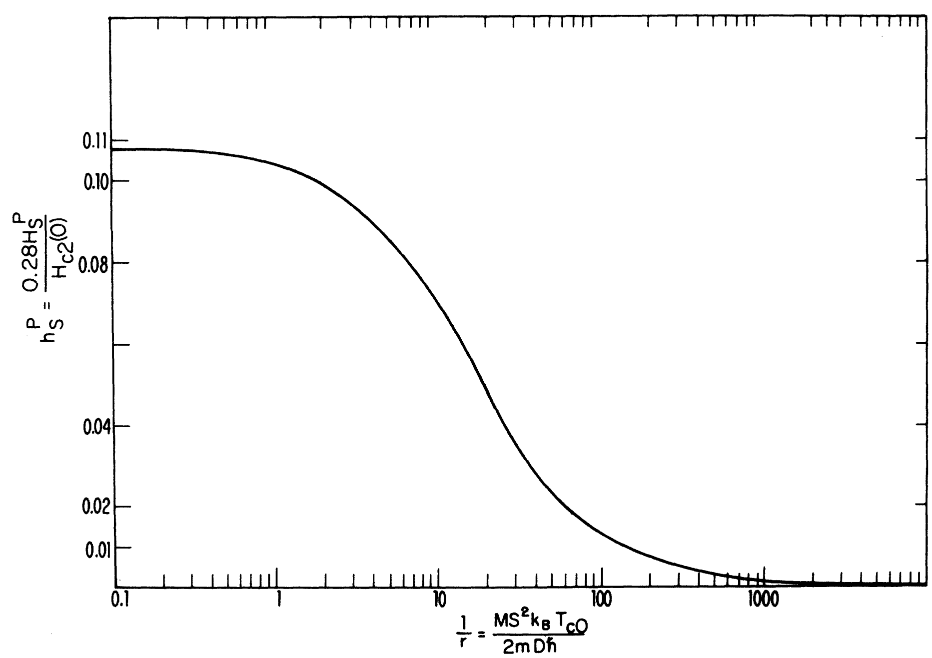

FIG. 8. Plot of $h_s^P = H_s^P/2\gamma' Hc_2(0)$ vs r^{-1}. H_s^P is defined to be the field that the layered magnetization equals $\frac{1}{2}$ the Prange value (in Fig. 1).

$$h=(T_{c0}/T)\,H/2\gamma' H_{c2}(0)\ , \tag{9}$$

where we have used the relation for $H_{c2}(0)$ in the dirty limit,

$$H_{c2}(0)=\pi T_{c0}/2\gamma' eD\ , \tag{10}$$

and $\gamma'=1.78$.

It can be shown that the term involving the sum over n gives a total contribution of order 5% or less of the magnetization. Therefore, for computational purposes, we may use the Ginzburg-Landau approximation for the terms in the infinite sum, whereas we shall use the full dirty limit propagator form for the other two terms. We shall hereafter refer to the magnetization obtained by dropping the infinite sum in Eq. (8) as $\mathfrak{M}_{\mathrm{major}}$. We note that at $H=0$, only the first term contributes to the susceptibility ($\chi=M/H$). It can also be shown that the Ginzburg-Landau limit for the entire expression in Eq. (8) gives the result of MT, if we drop the zero-point term (the $\frac{1}{2}$ in the frequency integral). As the Ginzburg-Landau limit is just the lowest term in the Taylor series expansion of the digamma functions for small $e(k_z)$, small ϵ_0, small ω, and low fields, we note that the correct value for the magnetization will always be less in magnitude than the approximate value. Since the results of MT gave a temperature dependence always greater than experiment,[5] we shall be careful to do the calculations without making this additional approximation on the two largest terms.

III. NEGLECT OF "ZERO-POINT" TERMS

To examine the "zero-point" terms for the two-dimensional and layered superconductors, we proceed analogously with the treatment of Maki[11] for the three-dimensional case. Since the ω integral for the zero-point part of Eq. (8) is divergent as it stands, we make use of the fact that at large ω, the diffusion constant becomes frequency dependent, in such a way as to cut the integral off at $\omega=\tau^{-1}$, where τ is the lifetime of the pair states. The term involving $f^{(1)}$ is then seen to be much larger than the other terms, as none of them are divergent without the cutoff. Keeping only this term, we may perform the ω integral, and obtain

$$\mathfrak{M}_{zp}\simeq-\frac{he}{12\pi^2}(4\pi T)\int\frac{dk_z}{2\pi}\,\mathrm{Re}\left\{\ln\left[\psi\left(\frac{1}{2}+\frac{-i\tau^{-1}+D(2eH+e(k_z))}{4\pi T}\right)-\psi\left(\frac{1}{2}-\frac{\epsilon_0}{4\pi T}\right)\right]\right.$$
$$\left.-\ln\left[\psi\left(\frac{1}{2}+\frac{D(2eH+e(k_z))}{4\pi T}\right)-\psi\left(\frac{1}{2}-\frac{\epsilon_0}{4\pi T}\right)\right]\right\}, \tag{11}$$

where $\mathfrak{M}_{zp}$ is the zero-point contribution to the magnetization.

Since $\tau T_{c0}\ll 1$, the two-dimensional zero-point magnetization is given by

$$\mathfrak{M}_{zp}^{2D}\simeq-\frac{heT}{3\pi d}\left\{\ln\ln\left(\frac{\gamma'}{\pi T_{c0}\tau}\right)-\ln\left[\psi\left(\frac{1}{2}+\frac{h}{2}\right)-\psi\left(\frac{1}{2}-\frac{\epsilon_0}{4\pi T}\right)\right]\right\}, \tag{12}$$

which we note is small for $H\ll H_{c2}(0)$, but varies weakly with temperature otherwise. This temperature dependence can be shown to be much weaker than the weakest temperature dependence from the other terms in the ω integral; if we look at the zero-field expression for the susceptibility, for example, the "zero-point" term diverges logarithmically as $T\to T_{c0}$, whereas the analogous term from the $(e^{\beta\omega}-1)^{-1}$ integral diverges as $(T-T_{c0})^{-1}$. Furthermore, for $H\gtrsim H_{c2}(0)$, the temperature dependence of the zero-point term becomes very weak, although the magnitude of the term is large. Since it is the temperature dependence of the magnetization that is measured, the zero-point term can also be neglected in two dimensions.

For the layered superconductors, we cannot perform the k_z integral exactly, but we note that for large s, the zero-point expression reduces to the two-dimensional form we have just calculated; and for small s, it reduces to the three-dimensional form, which was approximated by Maki[11] to show that it was weakly temperature dependent. The three-dimensional form does not diverge as $H\to 0$ and $T\to T_{c0}$, and away from T_{c0}, it can be shown to be as weak relative to the other (non-zero-point) terms just as the two-dimensional zero-point term was weak relative to the non-zero-point two-dimensional terms. An approximate calculation reveals that the zero-point term for layered superconductors can also be neglected for intermediate values of s.

IV. CALCULATION OF THE MAGNETIZATION

We now refer to Eq. (8) and examine the small contribution from the sum over n. Since the entire sum gives a contribution that can be shown to be of the order of 5% or less relative to the other two terms (e.g., in the Ginzburg-Landau approximation), and since the temperature dependence of these terms is stronger than for the other two terms, they will be even much less than 5% for high temperatures. We are therefore justified in approximating the terms in this sum by an approximation that will make the integrals simple: the Ginzburg-Landau approximation. We refer to the contribution to the magnetization from the term with the infinite sum as $\mathfrak{M}_{\text{minor}}$. We list the contributions to the magnetization from the following terms:

$$\mathfrak{M}^{3D}_{\text{minor}} \simeq \left(\frac{4e}{\hbar c}\right)^{3/2}\left(\frac{MH}{m}\right)^{1/2}\left(\frac{V}{4\pi\beta}\right) \times\left[\frac{1}{3\pi}\int_0^\infty \frac{dz}{e^z-1}\,\mathrm{Im}\left(\sum_{n=1}^\infty \sum_{\substack{m=5\\ \text{odd}}}^\infty \frac{(m-3)(2m-5)!!\,(2\gamma+iz/2\pi h+3n/m)}{2^m(2m-4)!!\,(n+\gamma+iz/4\pi h)^{m-1/2}}\right)\right], \tag{13}$$

$$\mathfrak{M}^{2D}_{\text{minor}} \simeq \frac{4eV}{4\pi\beta d\hbar c}\left[\frac{4}{3\pi}\int_0 \frac{dz}{e^z-1} \times\mathrm{Im}\left(\sum_{n=1}^\infty \sum_{\substack{m=5\\ \text{odd}}}^\infty \frac{(m-3)(n/m+\gamma+iz/4\pi h)}{2^m(n+\gamma+iz/4\pi h)^m}\right)\right] \tag{14}$$

$$\mathfrak{M}^{\text{layered}}_{\text{minor}} \simeq \frac{4eV}{4\pi\beta s\hbar c}\left\{\frac{2}{3\pi}\int_0^\infty \frac{dz}{e^z-1} \times\mathrm{Im}\left[\sum_{n=1}^\infty \sum_{\substack{m=3\\ \text{odd}}}^\infty \frac{m-1}{t_n^{m+1}} \times\left(P_m(z_n)-\frac{2n(m+1)P_{m+1}(z_n)}{(m+2)t_n}\right)\right]\right\}, \tag{15}$$

where P_m is the mth Legendre polynomial and t_n and z_n are given by

$$t_n = 2\,[(n+\gamma+iz/4\pi h+\sigma)^2-\sigma^2]^{1/2}, \tag{16}$$

$$z_n = 2(n+\gamma+iz/4\pi h+\sigma)/t_n, \tag{17}$$

$$\sigma = m/Ms^2\,2eH, \tag{18}$$

$$\gamma = \epsilon_0/4eHD, \tag{19}$$

and where ϵ_0 is given by Eq. (4). Note that we have not made the Ginzburg-Landau approximation for ϵ_0, although we did make it for the H, k_z, and ω dependence of the digamma functions. Thus, this approximation is quite reasonable.

The "major" terms are given by

$$\mathfrak{M}^{3D}_{\text{major}} = \left(\frac{4e}{\hbar c}\right)^{3/2} V\left(\frac{MH}{m}\right)^{1/2}\frac{1}{2\pi^2\beta}\int_0^\infty \frac{dz}{e^z-1}\,\frac{(h)^{1/2}}{12}\int_{-\infty}^\infty \frac{dk_z}{2\pi} \times\mathrm{Im}\left[\left(1-h\frac{\partial}{\partial h}\right)\left(\frac{\psi^{(1)}[(h+1)/2+iz/4\pi+k_z^2]}{\psi[(h+1)/2+iz/4\pi+k_z^2]-\psi(\frac{1}{2}-\epsilon_0/4\pi T)}\right)\right], \tag{20}$$

$$\mathfrak{M}^{2D}_{\text{major}} = \left(\frac{4e}{\hbar c}\right) V\left(\frac{1}{2\pi^2\beta d}\right)\int_0^\infty \frac{dz}{e^z-1}\left(\frac{h}{12}\right)\mathrm{Im}\left[\left(1-h\frac{\partial}{\partial h}\right) \times\left(\frac{\psi^{(1)}[(h+1)/2+iz/4\pi]}{\psi[(h+1)/2+iz/4\pi]-\psi(\frac{1}{2}-\epsilon_0/4\pi T)}\right)\right], \tag{21}$$

$$\mathfrak{M}^{\text{layered}}_{\text{major}} = \frac{(4e/\hbar c)V}{4\pi\beta s}\left(\frac{2}{\pi}\right)\int_0^\infty \frac{dz}{e^z-1}\left(\frac{h}{12}\right)\int_{-\pi}^{\pi}\frac{dk_z}{2\pi} \times\mathrm{Im}\left[\left(1-h\frac{\partial}{\partial h}\right)\left(\frac{\psi^{(1)}[(h+1)/2+iz/4\pi+(r/4\pi)(1-\cos k_z)]}{\psi[(h+1)/2+iz/4\pi+(r/4\pi)(1-\cos k_z)]-\psi(\frac{1}{2}-\epsilon_0/4\pi T)}\right)\right]; \tag{22}$$

here $r=2mD/Ms^2T$. The magnetizations used in the numerical work were just the "major" plus the "minor" magnetizations.

V. RESULTS AND DISCUSSION

To get a crude idea of how the tight-binding model for layered superconductors compares with two- and three-dimensional systems, we shall perform the calculations for two dimensions and layered systems within the spirit of Prange.[12] That is, we shall take the Ginzburg-Landau limit, and replace $(e^{\beta\omega}-1)^{-1}$ by $(\beta\omega)^{-1}$ and take the residue from the $\omega=0$ pole only (static limit). The expressions for these approximations are given below, and we note that the three-dimensional expression is formally identical with the result of Prange, multiplied by the anisotropy factor $(M/m)^{1/2}$:

$$f_{3D}(\gamma,\sigma)=f_p(\gamma)=\frac{1}{24}\left(\frac{\gamma+\frac{3}{4}}{(\gamma+\frac{1}{2})^{3/2}}+4\sum_{n=1}^{\infty}\sum_{\substack{m=5\\ \text{odd}}}^{\infty}\frac{(m-3)(2m-5)!!(2\gamma+3n/m)}{2^m(2m-4)!!(n+\gamma)^{m-1/2}}\right), \tag{24}$$

$$f_{2D}(\gamma,\sigma)=\left(\frac{\sigma}{2}\right)^{1/2}\left[\frac{1}{12}\left(\frac{\gamma+1}{(\gamma+\frac{1}{2})^2}\right)+\frac{2}{3}\sum_{n=1}^{\infty}\sum_{\substack{m=5\\ \text{odd}}}^{\infty}\frac{(m-3)(n/m+\gamma)}{[2(n+\gamma)]^m}\right], \tag{25}$$

$$f_{\text{layered}}(\gamma,\sigma)=\tfrac{1}{12}(\tfrac{1}{2}\sigma)^{1/2}\Big[[(\gamma+\sigma+\tfrac{1}{2})^2-\sigma^2]^{-1/2}+\tfrac{1}{2}(\gamma+\sigma+\tfrac{1}{2})[(\gamma+\sigma+\tfrac{1}{2})^2-\sigma^2]^{-3/2}+4\sum_{n=1}^{\infty}\sum_{\substack{m=3\\ \text{odd}}}^{\infty}\frac{m-1}{t_n^{m+1}}\left(P_m(z_n)-\frac{2n(m+1)}{(m+2)t_n}P_{m+1}(z_n)\right)\Big], \tag{26}$$

where

$$\mathfrak{M}=-(4e/\hbar c)^{3/2}(MH/m)^{1/2}(V/4\pi\beta)f(\gamma,\sigma) \tag{27}$$

and

$$t_n=2[(\gamma+\sigma+n)^2-\sigma^2]^{1/2}, \tag{28}$$

$$z_n=2(\gamma+\sigma+n)/t_n. \tag{29}$$

The results of numerical work at T_{c0} are given in Fig. 1. Note that $f_p(0)=0.09133$ and $f_{2D}(0,\sigma)=0.34589(\frac{1}{2}\sigma)^{1/2}$. Note that for small magnetic fields, the tight-binding model for layered superconductors approaches the Prange result, and at large magnetic fields, it approaches the "two-dimentional Prange" result, and passes smoothly from one extreme to the other. The transition region is for $\sigma=b^{-1}\sim 0.1$.

The expressions for the magnetizations in the limit $H\to 0$ can be computed exactly by keeping only the first terms of Eqs. (20)–(22), omitting the terms involving the derivative with respect to h, since those terms are negligible as $h\to 0$. Gollub, Beasley, and Tinkham[13] (GBT), who first performed experiments on dirty lead-thallium alloys, have performed an extrapolation of their data to zero magnetic field, and the temperature dependence of their extrapolated data is compared with the theory [numerical integration of the first term of Eq. (20)] in Fig. 2. Also plotted in Fig. 2 is the zero-field temperature dependence in the Ginzburg-Landau approximation, and the simple Schmid[4] theory. The deviations of the theory from experiment are seen to be large away from T_{c0}. We attribute this to the same effects that give the rather poor agreement with the low-field magnetization at T_{c0} in Fig. 3, although it must be admitted that the theory described here is also least reliable at very large $(T-T_c)/T_c$.

In Fig. 3, we have plotted the field dependence for dirty three-dimensional superconductors, and have compared our calculation with the calculations of MT and Prange, and the experiment of GBT. We note that the low-field agreement is rather poor, and the high-field agreement is only fair, but significantly better than the approximate calculation of MT. In Fig. 4, we have plotted the temperature dependence of the three-dimensional materials, and have again compared our calculations with those of MT and the experiments of GBT. The $H/H_s=0.3$ agreement is seen to be excellent; the $H/H_s=5$ agreement is far less quantitative, but it is better than that of the MT calculation. The quantity H_s is the field at which the magnetization is $\frac{1}{2}$ the static value. In Figs. 5 and 6, we have plotted the field and temperature dependences of two-dimensional thin films, for which H_s is found to be very nearly $H_{c2}(0)$. However, at this time there are no experimental data with which to compare our calculations.

The field dependence of layered superconductors for three values of the parameter r is given in Fig. 7. We note that the curves all approach the Prange value as $H\to 0$, and they approach the respective two-dimensional curves for large H. We note further that for $r\gtrsim 1$, that the layered superconductors exhibit three-dimensional field behavior, whereas for $r\ll 1$, the magnetization at large fields is suppressed. We have plotted h_s^p

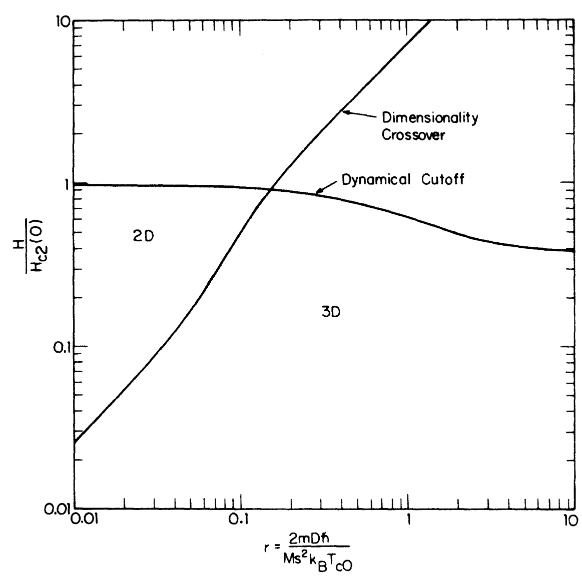

FIG. 9. Plot of $H_s/H_{c2}(0)$ and $H_D/H_{c2}(0)$, the fields at which the layered magnetization reaches $\frac{1}{2}$ the static value (see Fig. 1) and where the behavior crosses over from three- to two-dimensional behavior. H_D is defined to be the field where the two-dimensional magnetization equals the three-dimensional magnetization.

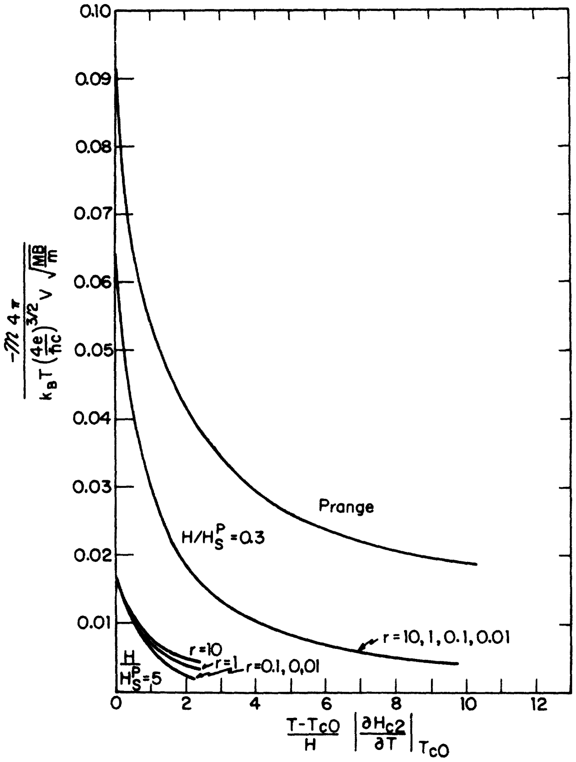

FIG. 10. Plot of the temperature dependence of layered superconductors for $H/H_s^P = 0.3$ and $H/H_s^P = 5$. (H_s^P is the field at which the magnetization is $\frac{1}{2}$ the Prange value.) Plotted are several curves for different r.

[defined to be that field where the layered superconductors approach $\frac{1}{2}$ the Prange value, divided by $2\gamma' H_{c2}(0)$] as a function of r^{-1} in Fig. 8. We note that for $r \simeq 0.01$, h_s^p is an order of magnitude less than the three-dimensional value.

To separate out the suppression of the magnetization due to dynamics from that due to dimensionality, we have plotted the dynamical cutoff field H_s and the dimensional crossover field H_D as a function of r in Fig. 9. We have defined H_s to be the field at which the magnetization equals $\frac{1}{2}$ the static value given by Eq. (26) and plotted in Fig. 1, and H_D is the field at which the two- and three-dimensional magnetizations are equal [e.g., in Fig. 1 the static magnetizations are equal at $b \sim 7.1$]. We therefore expect essentially two-dimensional behavior for $H > H_D$, and essentially three-dimensional behavior for $H < H_D$. Note that for $r \gtrsim 1$, the magnetization remains three-dimensional until far beyond the dynamical cutoff, but for $r \sim 0.01$, there are large regions of two-dimensional behavior. This can also be seen from Fig. 7.

Finally, in Fig. 10 we compare the temperature dependence of the magnetization for various values of r, and for $H/H_s^p = 0.3$ and 5. The quantity H_s^p is defined to be the field at which the magnetization is $\frac{1}{2}$ the three-dimensional static value, and is in general a function of r [see Fig. 8, where $H_s^p = 2\gamma' h_s^p H_{c2}(0)$]. We observe, however, that the $H_s/H_s^p = 0.3$ curve is very nearly independent of r, and comparison with Figs. 4 and 6 shows that it is three dimensional in nature. The $H_s/H_s^p = 5$ curve, however, is r dependent, and we observe that for $r = 10$ (and greater), it agrees with the three-dimensional theory in Fig. 4, whereas for $r = 0.1$ and less, it is two dimensional. For $r = 1$, it is apparently in the transition region. From Fig. 9, we note that for $r \lesssim 0.3$, we expect three-dimensional behavior for $H/H_s^p = 0.3$ and two-dimensional behavior for $H/H_s^p = 5$. For $r = 10$, we expect three-dimensional behavior for both curves. However, for $r = 1$, the $H/H_s^p = 5$ curve should be near the dimensionality crossover.

Preliminary experimental investigations by Foner[7] on TaS_2 (pyridine)$_{1/2}$ indicate that $H_{c2}(0)$ for applied fields perpendicular to the layers is of the order of 4 kG, which, from the expression for the dirty limit $H_{c2}(0)$, gives us an $E_F \tau \simeq 20$. This is within the allowable limits for the theory to be valid (although it is just barely within the allowable limits). Thus, the layered superconductors that have so far been made, appear to be extremely dirty. Prober, Beasley, and Schwall[14] have observed that the temperature dependence of one sample of $TaS_2(Py)_{1/2}$ at low magnetic fields was essentially identical with the temperature dependence of the three-dimensional alloy Pb-5-at.% Tl.[13] This is in agreement with the theory,[15] but it would be good to have more experimental data on thin films and layered superconductors. It is interesting to point out that if r were made of order 0.01, by using materials that have a larger layer spacing, or are more anisotropic, we would obtain a material that exhibited primarily two-dimensional-field behavior. We note that TaS_2 intercalated with octadecylamine has been shown to have a layer spacing of ~60 Å. If the mass ratio decreases exponentially with s, as expected in materials without too many shorts, these small values of r should be easily attainable experimentally.

Note added in proof. It has been brought to our attention by Gerhardts that the curve in Fig. 8 should have a bump in it, centered at $r \sim 1$. Careful reexamination of our computer calculations indicates that this is indeed the case, although this does not change the results qualitatively. For a curve that is more precise in this region of r, see Ref. 8.

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[1](a) F. R. Gamble, F. J. DiSalvo, R. A. Klemm, and T. H. Geballe, Science **168**, 568 (1970). (b) T. H. Geballe, A. Menth, F. J. DiSalvo, and F. R. Gamble, Phys. Rev. Lett. **27**, 314 (1971).

[2]J. Lawrence and S. Doniach, in *Proceedings of the Twelfth International Conference on Low-Temperature Physics*, edited by Eizo Kanda (Academic of Japan, Kyoto, 1971), p. 361.

[3]T. Tsuzuki, J. Low Temp. Phys. **9**, 525 (1972).

[4]A. Schmid, Phys. Rev. **180**, 527 (1969).

[5]K. Maki and H. Takayama, J. Low Temp. Phys. **5**, 313 (1971).

[6]D. Prober (private communications).

[7]S. Foner, Bull. Am. Phys. Soc. **17**, 289 (1972); and private communications.

[8]P. Gerhardts and S. Doniach, Bull. Am. Phys. Soc. **18**, 385 (1973); and report of work prior to publication.

[9]P. A. Lee and M. G. Payne, Phys. Rev. B **5**, 923 (1972).

[10]J. Kurkijärvi, V. Ambegaokar, and G. Eilenberger, Phys. Rev. B **5**, 868 (1972).

[11]K. Maki, Phys. Rev. Lett. **30**, 648 (1973).

[12]R. E. Prange, Phys. Rev. B **1**, 2349 (1970).

[13]J. P. Gollub, M. R. Beasley, and M. Tinkham, Phys. Rev. Lett. **25**, 1646 (1970); J. B. Gollub, M. R. Beasley, R. Callarotti, and M. Tinkham, Phys. Rev. B **7**, 3039 (1973).

[14]D. E. Prober, M. R. Beasley, and R. E. Schwall, in a more complete interpretation of the data reported in the Proceedings of the Thirteenth International Conference on Low Temperature Physics, Boulder, Colo., 1972 (unpublished).

[15]For $TaS_2(\text{pyridine})_{1/2}$ we estimate $r \sim 1$ using Foner's critical-field data (Ref. 7). For this value of r, three-dimensional behavior is expected over the limited temperature range $(T-T_c)/T_c \leq 0.2$ for which reliable data have been obtained.