Theory of the upper critical field in antiferromagnetic superconductors

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We compute the temperature T dependence of the upper critical field $H_{c2}(T)$ in antiferromagnetic (AF) superconductors. Using a strong-coupling formalism we explicitly treat the effects of the molecular field H_Q, inelastic and elastic spin-fluctuation scattering and magnetic as well as nonmagnetic impurities. A sum rule is used to relate the T dependence of H_Q to that of the spin-fluctuation scattering. The decreased pair breaking observed below the Néel temperature in $SmRh_4B_4$ and the increased pair breaking seen in the AF Chevrel compounds will both occur in our theory for a reasonable choice of parameters. For larger values of the dimensionless spin-exchange coupling constant $N(0)J^{cf}$, spin-fluctuation-scattering effects dominate over those of H_Q and decreased pair breaking is observed below T_N. For smaller values of the coupling constant, the converse is true. Impurity scattering is treated in a self-consistent fashion. As a consequence, the molecular field H_Q is altered by nonmagnetic impurities. This leads to important pair-breaking effects in H_{c2}. A physical manifestation of this pair breaking is a qualitative change in the shape of the H_{c2} versus T curve, as nonmagnetic impurities are added. We give detailed predictions for the expected effects of these impurities on H_{c2} which can be tested experimentally.

I. INTRODUCTION

It is now well established that superconductivity and antiferromagnetic order coexist in numerous ternary alloys. The Chevrel compounds, for example, $GdMo_6S_8$, $TbMo_6S_8$, and $DyMo_6S_8$, become antiferromagnetic at a temperature T_N which is below the superconducting transition temperature T_c. The rhodium-boride compound $SmRh_4B_4$ exhibits antiferromagnetic (AF) order at a T_N which is also less than the corresponding T_c. The reason that superconductivity coexists with antiferromagnetism is, in large part, due to the fact that the electrons which mediate the indirect Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction between the (magnetically ordered) localized rare-earth spins are distinct from those which undergo the superconducting pairing (which are associated with the transition elements in the ternary compounds). One major effect of the rare-earth ions is to create a (periodic) molecular field H_Q which can decrease the effective conduction-electron density of states in some regions of the Fermi surface. Here, Q is the wave vector of the AF order. In addition these localized electrons will break Cooper pairs by spin-fluctuation scattering and thereby further weaken superconductivity.

Many theoretical[1-6] and experimental[7-10] studies of these systems have focused on the temperature dependence of the upper critical field H_{c2}. Experiments[7-10] indicate that $H_{c2}(T)$ deviates markedly from the usual behavior[11] (of dirty superconductors) in the vicinity of T_N and below. The Chevrel compounds exhibit enhanced pair breaking below T_N, that is, they show a decrease in H_{c2}. By contrast, $SmRh_4B_4$ shows decreased pair breaking at the Néel temperature, therefore H_{c2} increases with respect to the value extrapolated from high T. A number of theoretical approaches[2,3,5] have been able to obtain reasonable agreement with experimental $H_{c2}(T)$ measurements in the AF superconductors. This is unexpected since these theories are based on different physical models and assumptions which are, in some cases, incompatible. There has yet been no sufficiently complete theory for treating simultaneously all of the physical effects known to be important in the ternary compounds. It is the purpose of this paper to present the most detailed calculation yet of $H_{c2}(T)$ which includes simultaneously (i) the effects of the antiferromagnetic molecular field H_Q, (ii) inelastic spin-fluctuation scattering, and (iii) nonmagnetic as well as magnetic impurity effects. We also discuss spin-orbit scattering and paramagnetic effects arising from the applied field. Our paper is based on previous work by Nass *et al.*[12] These authors studied inelastic spin-fluctuation and molecular-field effects in AF superconductors in the absence of electromagnetic fields.

The first microscopic calculation of H_{c2} in an AF superconductor is due to Maekawa and Tachiki.[1] These authors set up a general strong-coupling formalism for the superconducting Green's function in the presence of electromagnetic fields and inelastic spin-fluctuation scattering. This general theory, however, was applied to the paramagnetic phase of AF (and ferromagnetic) superconductors so that the molecular field H_Q was taken to be zero. Furthermore, all spin-fluctuation-scattering processes were ultimately treated as elastic; they could therefore be combined with the phonons into an effective coupling constant $\tilde{g}$. Machida, Nokura, and Matsubara[3] improved upon the work of Ref. 1 by including as a perturbative effect a nonzero H_Q. This approximate treatment of the molecular field, along with the assumption of elastic spin-fluctuation scattering led to a new renormalized

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coupling constant $\bar{g}$. As in Ref. 1, the usual weak-coupling expressions for H_{c2} and other quantities could then be directly applied to the AF superconductors.

Ramakrishnan and Varma[4] correctly pointed out that a treatment of spin-fluctuation scattering must include inelastic as well as elastic processes; these can be included in a calculation of H_{c2} using a strong-coupling formalism.[13] It was asserted[4] that in dirty systems, molecular-field effects were relatively unimportant and that the major effect of the onset of magnetic ordering on superconductivity comes through the spin-fluctuation coupling $\alpha^2F^{\rm mag}(\omega)$. For a spherical Fermi surface, if $H_Q=0$, the freezing out of spin fluctuations at and below T_N leads to *decreased* pair breaking in the antiferromagnetic state.[1,3] This is consistent with the behavior observed in some but not all of the ternary compounds. The authors in Ref. 4 argued that increased pair breaking could be obtained if the Fermi surface was nonspherical and, in particular, if the contribution of the local-spin susceptibility $\chi(q,\omega_n-\omega_m)$ had increased weight at $q=Q$, due to a joint density-of-states effect. Therefore, it was argued, deviations from a spherical Fermi surface were essential in order for magnetic order to suppress H_{c2} at T_N and below.

Finally, a very different calculation of $H_{c2}(T)$ was presented by Zwicknagl and Fulde,[5] who asserted that the superconducting pairing in AF superconductors does not involve time-reversed electrons, but rather takes place between magnetic quasiparticle states. In Ref. 5, H_{c2} was calculated using the strong-coupling formalism of Eilenberger[14] and Eliashberg[13] and considering only the phonon part of the interaction; spin-fluctuation effects were neglected. It was argued that in the new pairing state, the appropriate phonon spectral function $\alpha^2F^{\rm ph}(\omega)$ was modified because the pairing takes place between the magnetic quasiparticles. It should be stressed that the actual phonon propagator was assumed to be unaffected by the AF order.

Comparison between theory and experimental H_{c2} measurements was made in Refs. 3 and 5. In these previous works, the agreement with the data was quite satisfactory. Nevertheless, because none of the above theories has included a study of the very dramatic impurity effects and because all of these theories have obvious weaknesses, we are motivated to recalculate $H_{c2}(T)$. Machida *et al.*[3] claimed that the molecular field enters into the linearized gap equation in powers of H_Q/E_F. Therefore, it was treated perturbatively. However, it has been pointed out[12] that this perturbation approach is incorrect because the relevant parameter is H_Q/ω_c, where ω_c is related to the superconducting cutoff or Debye frequency. This ratio is not particularly small. As a consequence the molecular field cannot be incorporated into an effective coupling constant as was done in Ref. 3. Furthermore, there is a delicate interplay between the effects of impurities and the molecular field, which was ignored in Ref. 3. A renormalized field H_Q must be self-consistently calculated in the presence of magnetic and nonmagnetic impurities, just as the gap Δ and frequency ω are renormalized in dirty superconductors.[11] While Ramakrishnan and Varma[4] improved upon the work of Machida *et al.*[3] by allowing the spin-fluctuation scattering to be inelastic, they did not explicitly include the molecular field H_Q. At present, the work of Zwicknagl and Fulde[5] is controversial since it involves the ansatz that the pairing is between magnetic quasiparticles. It should be noted that in an earlier paper Machida *et al.*[15] also considered the possibility of pairing between non-time-reversed states. However, subsequent work by Nass *et al.*[16] and Machida *et al.*[3] showed that for three-dimensional weak-coupling superconductors the dominant pairing is of the Cooper type. Unlike some calculations, no approximations were used to handle the three dimensionality in the work of Ref. 16.

Our formalism for calculating H_{c2} is based on strong-coupling theory. However, our H_{c2} equation differs from that considered in Ref. 4 because of the presence of the renormalized molecular field $\tilde{H}_Q(\omega)$. This field is obtained by solving a pair of coupled equations for the renormalized frequency $\tilde{\omega}(\omega)$ and field $\tilde{H}_Q(\omega)$ in the presence of impurities as well as spin fluctuations. For computational facility we use the quasi-three-dimensional approximation of Bilbro and McMillan;[17] we assume that the molecular field is nonzero only in certain regions of the Fermi surface and it is zero elsewhere. In the regions where $H_Q\neq 0$ the energy dispersion curves are assumed to be one dimensional (1D), and hence $\epsilon_k=-\epsilon_{k+Q}$. Because this approach represents a composite of a nonmagnetic BCS superconductor and a 1D coexistent AF superconductor, many of our numerical results will be presented for a one-dimensional system. The effects of (quasi-) three-dimensionality (3D) can be readily extrapolated from our 1D calculations. The quasi-3D approach is itself clearly oversimplified. However, previous numerical (weak-coupling) studies[12] which have compared this with a more exact calculation of 3D Fermi-surface integrals indicate that this approximation is not unreasonable.

Our model for the dynamic spin-fluctuation propagator is similar to that used in Refs. 4 and 12. As in these previous works we impose the usual sum rule which relates the magnetic order parameter $\langle S_Q\rangle$ to the susceptibility χ. This implies that the temperature dependences of H_Q and of χ are not independent, and represents an important departure from the work of Machida *et al.*[3] The molecular field H_Q is given by $nJ^{cf}\langle S_Q\rangle$, where n is the number of rare-earth ions per unit cell and J^{cf} is the conduction-electron local (f) spin exchange. χ enters the H_{c2} equation multiplied by a term $n(J^{cf})^2N(0)$ [where $N(0)$ is the density of states at the Fermi energy]. Therefore, the relative importance of spin-fluctuation and molecular-field effects depends on the size of the parameter $N(0)J^{cf}$. This parameter may vary considerably from one ternary system to another.

It should be stressed that it is the relative importance of these two mechanisms which determines the overall qualitative characteristics of the H_{c2}-versus-T curves. If the molecular field is relatively larger than the spin-fluctuation coupling, then $H_{c2}(T)$ exhibits a pronounced dip at T_N. If the converse is true then $H_{c2}(T)$ remains monotonic in temperature, but the magnitude of the slope jumps at T_N.

Among the more interesting consequences of this calculation is our observation that because nonmagnetic impur-

ities are pair breaking, they can affect the *shape* of the H_{c2}-versus-T curves. When nonmagnetic impurities are added, the role of the molecular field is diminished; a dip at T_N in H_{c2} may thus decrease in magnitude as the system is made dirtier. Our impurity calculations are in the same spirit as those which deal with the usual (nonmagnetic) superconductors. Thus we do not consider impurity effects on the spin-fluctuation propagator. While our quantitative predictions are obviously not accurate, it will be important for future H_{c2} experiments to test these qualitative effects and to clearly establish the pair-breaking nature of nonmagnetic impurities in the AF superconductors.

II. GENERAL THEORY

A. Derivation of equation for H_{c2}

For a ternary superconductor in an electromagnetic field we use the same model Hamiltonian as that considered by Maekawa and Tachiki.[1] However, in order to treat the effects of the AF molecular field, we add and subtract mean-field contributions. We write

$$\mathcal{H}=\mathcal{H}^0+\mathcal{H}^{\rm int}+\mathcal{H}^A+\mathcal{H}^{\rm imp}\ , \tag{2.1}$$

where in terms of the creation and annihilation operators c_p, $c_p^\dagger$, etc.,

$$\mathcal{H}^0=\sum_{p,\sigma}\epsilon_p c_{p\sigma}^\dagger c_{p\sigma}+\sum_{p,\sigma}\sigma(H_Q c_{p+Q\sigma}^\dagger c_{p\sigma}+H c_{p\sigma}^\dagger c_{p\sigma})\ , \tag{2.2a}$$

$$\mathcal{H}^{\rm int}=g\sum_\sigma\int d^3x\,\phi(x)c_\sigma^\dagger(x)c_\sigma(x)-\frac{J^{cf}}{N}\sum_{R_i}\int d^3x\,\delta(x-R_i)\{\tilde{S}^z(R_i)[c_\uparrow^\dagger(x)c_\uparrow(x)-c_\downarrow^\dagger(x)c_\downarrow(x)]$$
$$+S^+(R_i)c_\downarrow^\dagger(x)c_\uparrow(x)+S^-(R_i)c_\uparrow^\dagger(x)c_\downarrow(x)\}\ , \tag{2.2b}$$

and

$$\mathcal{H}^A=\sum_\sigma\int d^3x\ c_\sigma^\dagger(x)\left[-\frac{\hbar^2}{2m}\left[\vec{\nabla}-\frac{ie}{\hbar c}\vec{A}\right]^2+\frac{\hbar^2}{2m}\nabla^2\right]c_\sigma(x)\ . \tag{2.2c}$$

Here, $\mathcal{H}^{\rm imp}$ represents the usual[11] interaction between the conduction electrons and magnetic as well as nonmagnetic impurities and will not be written here. In Eqs. (2.2), ϵ_p represents the conduction-electron kinetic energy before the effects of the new magnetic Brillouin zone are included. The field H appearing in Eq. (2.2a) is given in terms of the applied field H_0 as $H=H_0(1+\chi_0 J^{cf})$ where χ_0 is the uniform static spin susceptibility of the rare-earth spins $\chi_0\equiv\langle S^z\rangle_0/H_0$. The parameters in the molecular field, $H_Q\equiv n\langle S^z{}_Q\rangle J^{cf}$, were discussed in Sec. I. This field will be taken to be a phenomenological parameter in most of our calculations. The two terms in $\mathcal{H}^{\rm int}$ represent, respectively, the interaction of the phonons (g is the phonon coupling constant and ϕ is the phonon field operator) and the conduction electrons and that of the rare-earth spins and conduction electrons, respectively. We define $\tilde{S}^z\equiv S^z-\langle S^z\rangle$. Finally, $\mathcal{H}^A$ represents the orbital coupling between the electromagnetic field and the conduction electrons. The vector potential $\vec{A}$ appearing in Eq. (2.2c) is assumed to be unaffected by the internal molecular field of the rare-earth spins. This approximation is valid for moderately large Q ($\gtrsim\xi^{-1}$), which seems to be satisfied[18] for the Chevrel compounds. We may also include a spin-orbit contribution to $\mathcal{H}$. Presumably, this interaction is rather strong in most of the ternary superconductors. Its effect is primarily to weaken the Pauli paramagnetic term in Eq. (2.2a). Therefore, in our calculations we will ignore the paramagnetic splitting of the conduction-electron bands. Furthermore, we assume that the superconductor is sufficiently dirty, so there are no explicit dependences on the spin-orbit-scattering lifetime. In the same spirit, the H dependence of the phenomenological parameter H_Q and of the spin fluctuations is neglected. These assumptions are expected to be valid providing also that $|\langle S_Q^z\rangle| \gg |\langle S_0^z\rangle|$, i.e., the mean-field-induced ferromagnetic ($Q=0$) component of the rare-earth spins is much smaller than the AF order parameter. In sufficiently large applied fields H_0, and/or sufficiently close to T_N, this inequality will clearly break down. Neutron experiments[18] indicate that for all $H\le H_{c2}$, and for $T<T_N$, the ferromagnetic component is relatively unimportant in $TbMo_6S_8$, but not in $DyMo_6S_8$. Therefore, our theory cannot be directly applied to this last compound, but should be applicable to $TbMo_6S_8$ and probably[19] to $GdMo_6S_8$ and $SmRh_4B_4$ as well, except for $T\approx T_N$. While it is not conceptually difficult to include paramagnetic effects in our equations, they do complicate their numerical solution. For this reason we will ignore them at present. Finally, we note that we have not considered crystal-field effects in $\mathcal{H}$. In addition, for simplicity, we will assume that the Fermi surface (when $H_Q=0$) is spherical. The effects of a nonspherical Fermi surface can be readily incorporated following Ref. 4.

A mean-field theory of an AF superconductor may be derived by adding to $\mathcal{H}^0$ a BCS-type electron-phonon interaction. The resulting Hamiltonian is readily diagonalized and the Green's functions may be calculated. In the presence of a vector potential $\vec{A}$ the normal-state Green's function $\tilde{G}$ is related to that obtained when $\vec{A}=0$ by the line integral

$$\tilde{G}(x,x';t-t')$$
$$\cong\tilde{G}^{A=0}(x-x';t-t')\exp\left[i\frac{e}{\hbar c}\int_x^{x'}d\vec{s}\cdot\vec{A}(\vec{s})\right]\ , \tag{2.3}$$

and therefore $\tilde{G}$ is shifted by a phase factor relative to the case $\vec{A}=0$. The above equation follows from the semiclassical approach, which is valid[20] when the superconductor is sufficiently dirty so that

$$\frac{e\hbar}{mc}H \ll k_B T+\hbar(\tau_1^{-1}+\tau_2^{-1}) , \tag{2.4}$$

where τ_1 and τ_2 correspond to the lifetime for scattering from nonmagnetic and magnetic impurities, respectively. This equation corresponds physically to the condition that the mean free path is sufficiently short so that the Lorentz force does not have time to alter the trajectory of the electrons between collisions.

The superconducting order parameter is defined as

$$\Delta(x;t)\propto \lim_{x\to x'}\langle T[c_\sigma(x;t)c_{-\sigma}(x';0)]\rangle . \tag{2.5}$$

It follows from Hartree-Fock theory and the standard[11] theory of weak impurity scattering that

$$\Delta(x;\omega_n)=\beta^{-1}\sum_q\sum_m\int d^3x'\lambda(q;\omega_n-\omega_m)\langle\tilde{G}^{A=0}(x-x';\omega_m)\tilde{G}^{A=0}(x-x';-\omega_m)\rangle_{\rm imp}\exp[i\vec{\pi}\cdot(\vec{x}-\vec{x}')]\Delta(x;\omega_m) , \tag{2.6}$$

where $\pi_i\equiv\partial_i-ieA_i/\hbar c$, ω_n is a Matsubara frequency, and $i=x$, y, or z. Here, $\langle\ \rangle_{\rm imp}$ denotes an impurity average, and

$$\lambda(q;\omega_n-\omega_m)=D(q;\omega_n-\omega_m)-\chi^{zz}(q;\omega_n-\omega_m)-\tfrac{1}{2}[\chi^{+-}(q;\omega_n-\omega_m)+\chi^{-+}(q;\omega_n-\omega_m)] , \tag{2.7}$$

where D and χ are the usual phonon and spin-fluctuation time-ordered propagators, respectively, multiplied by the appropriate coupling constants. Because we assume that the Fermi surface is spherical, λ will be averaged over q. It has been pointed out[4] that in some compounds there may be important q-dependent effects in λ associated with a nonspherical Fermi surface. We ignore these here in order to (i) isolate the competing effects of molecular-field and spin-fluctuation scattering, and (ii) because there is, as yet, no quantitative theory of the Fermi surfaces of the ternary superconductors. It follows from Eq. (2.6) that

$$\Delta(x;\omega_n)=\frac{1}{\beta V}\sum_m\int d^3x'\{K_{nm}(p,-p)+K_{nm}(p,-p+Q)e^{i\vec{Q}\cdot\vec{x}'}+K_{nm}(p,-p-Q)e^{-i\vec{Q}\cdot\vec{x}'}\}$$

$$\times\exp[i(\vec{p}-\vec{\pi})\cdot(\vec{x}-\vec{x}')]\Delta(x;\omega_m) , \tag{2.8}$$

where $\beta=(k_BT)^{-1}$, and the kernel

$$K_{nm}(p,p')=\sum_q\lambda(q;\omega_n-\omega_m)\sum_{p_1p_2p_3p_4}\langle\tilde{G}^{A=0}_{\sigma\sigma}(p_1,p_2;\omega_m)\tilde{G}^{A=0}_{-\sigma-\sigma}(p_3,p_4;-\omega_m)\rangle_{\rm imp}\delta_{p,p_1+p_3}\delta_{p',p_2+p_4} . \tag{2.9}$$

Here, we have used the fact that the only possible superconducting pairings are with $(p,-p)$ and $(p,-p-Q)$. The latter pairing corresponds to the order parameter Δ_Q in Ref. 16. Since this has been shown to be negligible[3,16] we will ignore the second and third terms in $\{\ \}$ in Eq. (2.8). Within the semiclassical approximation[20] [Eq. (2.3)] it then follows that

$$\Delta(x;\omega_n)=\beta^{-1}\sum_m K_{nm}(\pi,-\pi)\Delta(x;\omega_m) , \tag{2.10}$$

where $\pi=(\vec{\pi}\cdot\vec{\pi})^{1/2}$ and K is now a function of the operator π. In the 1D limit, the kernel may readily be evaluated using the normal-state Green's functions and

$$\tilde{G}^{A=0}(p,p;\omega_n)=\frac{\tilde{\omega}_n+\epsilon_p}{\tilde{\omega}_n^2+\epsilon_p^2+\tilde{H}_{Qn}^2} , \tag{2.11a}$$

$$\tilde{G}^{A=0}(p,p\pm Q;\omega_n)=\frac{\tilde{H}_{Qn}}{\tilde{\omega}_n^2+\epsilon_p^2+\tilde{H}_{Qn}^2} . \tag{2.11b}$$

In the absence of impurities and spin-fluctuation effects the renormalized quantities $\tilde{\omega}_n$ and $\tilde{H}_{Qn}$ are replaced by ω_n and H_Q, respectively. It may be readily verified that the resulting (two-component) nonrenormalized Green's function $G^{A=0}$ thus obtained corresponds to the Green's function for the Hamiltonian $\mathscr{H}^0$. Strong-coupling phonons, spin fluctuation, and impurity effects renormalize the frequency and molecular field as follows:

$$\tilde{\omega}_n=\omega_n+\beta^{-1}N(0)\sum_m\int_0^{2k_F}\frac{dq\,q}{2k_F^2}\{D(q;\omega_n-\omega_m)+\chi^{zz}(q;\omega_n-\omega_m)+\tfrac{1}{2}[\chi^{+-}(q;\omega_n-\omega_m)+\chi^{-+}(q;\omega_n-\omega_m)]$$

$$+(\hbar/2)(\tau_1^{-1}+\tau_2^{-1})\delta_{mn}\}\frac{\tilde{\omega}_m}{(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}} , \tag{2.12a}$$

and

$$\tilde{H}_{Qn}=H_Q-\beta^{-1}N(0)\sum_m\int_0^{2k_F}\frac{dq\,q}{2k_F^2}\{D(q;\omega_n-\omega_m)+\chi^{zz}(q;\omega_n-\omega_m)+\tfrac{1}{2}[\chi^{+-}(q;\omega_n-\omega_m)+\chi^{-+}(q;\omega_n-\omega_m)]$$
$$-(\hbar/2)(\tau_1^{-1}-\tfrac{1}{3}\tau_2^{-1})\delta_{mn}\}\frac{\tilde{H}_{Qm}}{(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}}\ . \tag{2.12b}$$

Substituting Eqs. (2.11) into (2.9) and performing a contour integral, it follows that

$$K_n(\pi,-\pi)\cong 2\pi N(0)\left[1-\frac{\tilde{H}_{Qn}^2}{\tilde{\omega}_n^2+\tilde{H}_{Qn}^2}\right]\tan^{-1}\left[\frac{v_F\pi}{2(\tilde{\omega}_n^2+\tilde{H}_{Qn}^2)^{1/2}}\right]\ . \tag{2.13a}$$

Assuming a solution $\Phi(x)$ exists for the eigenvalue equation

$$\pi^2(x)\Phi(x)=\alpha\Phi(x)\ , \tag{2.13b}$$

and that

$$\Delta(x;\omega_n)=\Phi(x)\Delta_n\ ,$$

one then arrives at a matrix equation,

$$\Delta_n=2\pi N(0)\beta^{-1}\sum_m\lambda(\omega_n-\omega_m)\frac{\Delta_m}{[1-\tilde{H}_{Qm}^2/(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)]^{-1}[2(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}+(e\hbar/c)v_F^2H_{c2}/6(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}]-\hbar(\tau_1^{-1}-\tau_2^{-1})}\ . \tag{2.14}$$

It can be shown that the eigenvalue $\alpha=eH_{c2}/\hbar c$ in Eq. (2.13b). Here, we have expanded $\tan^{-1}$ in Eq. (2.13a) to third order in its argument. Equation (2.14) represents the key equation of this section. While it was derived in the 1D limit, one may readily extend this equation to three-dimensional systems using the quasi-3D approximation of Bilbro and McMillan.[17] In this approximation, the term on the right-hand side of Eq. (2.14) is weighted by a factor γ (corresponding to the fraction of the Fermi surface which is strongly affected by the molecular field H_Q). An additional term, weighted by $1-\gamma$, is added to the right-hand side of Eq. (2.14). This term is obtained by setting $\tilde{H}_Q=0$ on the right-hand side of Eq. (2.14); this is the usual strong-coupling expression for H_{c2} in nonmagnetic superconductors. It may be verified that, in the limit $H=0$, Eq. (2.14) reduces to the (1D) gap equation for AF superconductors studied elsewhere.[16] In the limit $\tilde{H}_Q=0$, Eq. (2.14) is formally equivalent to the H_{c2} equation derived previously.[4] The presence of the molecular field leads to the additional complication (which has not previously been considered) that the renormalized frequency and field $\tilde{\omega}$ and $\tilde{H}_Q$ are now derived from coupled equations.

B. Elastic scattering: Impurity effects

In the limit in which the phonon-induced electron-electron interactions can be treated by the usual BCS weak-coupling approximation, and when inelastic spin-fluctuation effects are neglected, Eq. (2.14) can be written as

$$\ln\left[\frac{T}{T_{c0}}\right]=\int_0^\infty d\omega\tanh\left[\frac{\omega}{2}\right]\mathrm{Re}\left[\left[\beta\tilde{H}_Q(u^2-1)^{1/2}+\frac{\beta e\hbar v_F^2H_{c2}/c}{6\tilde{H}_Q(u^2-1)^{1/2}}-i\beta\hbar(\tau_1^{-1}-\tau_2^{-1})\right]^{-1}-1/\omega\right]\ . \tag{2.15}$$

Here, $u=\tilde{\omega}(\omega)/\tilde{H}_Q(\omega)$ and T_{c0} is the superconducting transition temperature with $H_Q=H=0$. This equation represents a "weak-coupling" approximation to Eq. (2.14). It follows from Eqs. (2.12) that u satisfies

$$\frac{\omega}{H_Q}=u\left[1-\frac{\hbar\tau_0^{-1}}{H_Q}\frac{1}{(1-u^2)^{1/2}}\right]\ . \tag{2.16}$$

Here, we have defined $\tau_0^{-1}\equiv\tau_1^{-1}+\frac{1}{3}\tau_2^{-1}$. In this weak-coupling limit, the two equations for the renormalized quantities $\tilde{\omega}$ and $\tilde{H}_Q$ can be expressed in terms of a single equation for the variable u. It should be noted that u is a complex quantity and that the square root must be taken at a fixed branch. Equation (2.16) is reminiscent of an analogous equation for the ratio of renormalized frequency to superconducting gap (also called u in the literature[11]), in superconductors containing magnetic impurities. The quantity u in Eq. (2.16) is related to the density of states in the normal antiferromagnet $N_N(\omega)$ as

$$N_N(\omega)=N(0)\mathrm{Im}\frac{u(\omega)}{[1-u^2(\omega)]^{1/2}}\ . \tag{2.17}$$

Thus, there is an analogy between the behavior of the gap in the superconducting density of states due to the superconducting order, and that in $N_N(\omega)$ due to the molecular field. As τ_0 decreases (due to nonmagnetic or magnetic impurities) the normal-state AF gap decreases. In a dirty

BCS superconductor, the superconducting gap weakens as τ_2 decreases. In each case, impurity levels fill in the gap in the density of states.

When the AF superconductor satisfies the inequality $\tau_0^{-1} \gg H_Q$, Eq. (2.16) may be solved to yield the closed-form expression

$$u(\omega)=\frac{\omega}{H_Q}\mp i\frac{\hbar\tau_0^{-1}}{H_Q}\ , \tag{2.18a}$$

with

$$\tilde{\omega}(\omega)=\omega\mp i\hbar(\tau_1^{-1}-\tau_2^{-1}) \tag{2.18b}$$

and

$$\tilde{H}_Q(\omega)=H_Q\frac{1\mp\hbar/2(\tau_1^{-1}-\frac{1}{3}\tau_2^{-1})}{\omega\mp\hbar/2(\tau_1^{-1}+\tau_2^{-1})}\ , \tag{2.18c}$$

where the upper (lower) signs correspond to positive (negative) frequency. In this limit, the impurity lifetime enters as a pure imaginary contribution to $\tilde{\omega}$. Since $|u(\omega)|^2 \gg 1$ for all $\omega \geq 0$, it follows that $H_Q(u^2-1)^{1/2} \approx \tilde{\omega}$, and the terms involving τ_1 cancel in Eq. (2.15). Therefore, nonmagnetic impurities are not pair breaking when $\tau_0^{-1} \gg H_Q$. It should be stressed that when this inequality is *not* satisfied nonmagnetic impurities will lead[12] to pair breaking similar to the behavior of magnetic impurities in nonmagnetic superconductors. However, when an AF superconductor is sufficiently dirty (so that the *normal* state is essentially "gapless") then Anderson's theorem[11] is reinstated. It has been shown[12] that, as additional nonmagnetic impurities are added, a gap reopens in the superconducting density of states $N_s(\omega)$, and $N_s(\omega)$ becomes BCS-like. It should be noted that impurity effects on the "bare" molecular field H_Q have not been explicitly treated in this analysis. It is expected that as τ_1 decreases, H_Q also decreases; this should act to reinforce all of the explicit effects of τ_1 discussed above.

These results for the τ_1 dependence of H_{c2} can also be restated in terms of the behavior of the superconducting coherence length $\xi=\alpha^{-1/2}$. It follows from Eq. (2.13b) that

$$H_{c2}(T)=\phi_0/\xi^2(T)\ , \tag{2.19}$$

where $\phi_0=\hbar c/2e$ is the unit of quantized flux. The pair-breaking effects of τ_1 arises through the dependence of ξ on τ_1. The coherence length depends on τ_1 through both the mean free path l, and for $H_Q \neq 0$, the transition temperature T_c. Both dependences are of opposite sign, so that ξ has a maximum when plotted as a function of $1/\tau_1$. With the use of Eq. (2.19) this implies that H_{c2} will initially decrease with increasing $1/\tau_1$; as $1/\tau_1$ increases further, H_{c2} goes through a minimum and then increases linearly. In our numerical studies we will assume that the superconductors are already sufficiently disordered, and therefore we only observe the increase in H_{c2} with increasing disorder.

When the inequality $H_Q \ll \tau_0^{-1}$ is not satisfied, Eq. (2.15) must be solved numerically. This is most readily done by converting it to a quartic equation and using iteration techniques. To obtain the proper root of this equation we require that as $\omega \to \infty$,

$$\tilde{\omega}(\omega) \to \omega + i\hbar(\tau_1^{-1}+\tau_2^{-1})\ ,$$

$$\tilde{H}_Q(\omega) \to H_Q\ .$$

After $u(\omega)$ is obtained, the resulting values are then substituted into Eq. (2.15) and H_{c2} is calculated using numerical integration techniques.

The resulting H_{c2}-versus-T curves are plotted in Fig. 1 for various τ_1^{-1} in units of ω_c for fixed $\tau_2^{-1}=10^{-5}\omega_c$. Here, ω_c is the Debye frequency. We chose the T dependence of H_Q to be of the form

$$H_Q(T)=H_Q(0)\left[1-\left[\frac{T}{T_N}\right]^{\nu}\right]\ , \tag{2.20}$$

where $2 \leq \nu \leq 4$ was deduced by a fitting to experimental[7,8] zero-field magnetization data. The functions for $\nu=2$ and 4 are plotted in the inset of Fig. 1. In the main part of the figure we use $\nu=2$. In addition, $H_Q(0)$ was taken to be $0.3\omega_c$ based on experimental considerations.[21] The τ_1 and τ_2 values are chosen in such a manner as to satisfy the "dirty" condition in Eq. (2.4). This justifies the use of the quasi-classical approximation. While our results are plotted for the 1D limit, the behavior of $H_{c2}(T)$ in three dimensions may be estimated by adding, to our 1D curves (properly weighted), the functional form[11] for H_{c2} obtained for (nonmagnetically ordered) dirty superconductors. In the cleanest system ($\tau_1=3.0\omega_c^{-1}$) there is reentrant behavior. The effect of the molecular field H_Q is so great that it completely destroys superconductivity for a range of temperatures. When $H_Q(T)$ approaches the saturation value, at low T superconductivity can redevelop. As the system becomes dirty the effects of H_Q

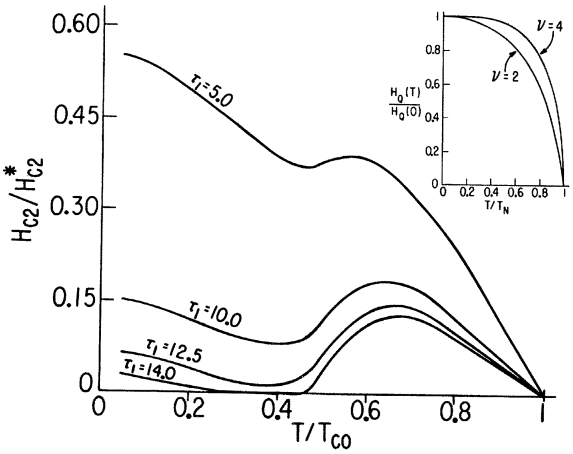

FIG. 1. Weak-coupling calculation of the temperature dependence of H_{c2} in units of $H^*_{c2}=c\omega_c^2/\hbar v_F^2 e$. Here, T_{c0} is the $H_Q=H=0$ superconducting transition temperature, and the nonmagnetic impurity scattering time τ_1 is varied in units of ω_c, the cutoff frequency which was chosen to be 100 K. In the inset is plotted the T dependence of the bare molecular field for the two exponents $\nu=2$ and 4.

are "screened" out so that the dip in H_{c2} at T_N is not as pronounced. The possibility of reentrant superconductivity was raised by Machida *et al.*[3,22] It should be noted, however, that in their work the effects of impurities were not treated self-consistently, and, therefore, H_Q was not renormalized. This renormalization appears to be extremely important and leads to considerable variation in the functional form of $H_{c2}(T)$. As will be stressed in Sec. III, a systematic experimental study of the dependence of $H_{c2}(T)$ on nonmagnetic impurity concentration is presently lacking, although this would be extremely important. It should finally be noted that in Fig. 1 we have ignored any shift in T_N as τ_1 varies. Presumably, T_N will decrease as τ_1 decreases. The effect on H_{c2} can readily be extracted from our numerical plots, since it primarily results in a rescaling of the temperature variable.

C. Models for the spin-fluctuation and phonon propagators: Inelastic effects

In order to solve Eq. (2.14) together with Eqs. (2.12) we need to adopt simple models for the phonon and spin-fluctuation propagators, called D and χ, respectively. For the phonons, we use the Einstein independent-oscillator model. For the spin fluctuations we adopt a simple form[4] which includes an Einstein-like model for the spin waves as well as longitudinal (elastic) modes. The magnon spectral weights which are chosen to be k independent are given by, for $T \leq T_N$,

$$\begin{aligned}\tfrac{1}{2}[B^{+-}(\omega)+B^{-+}(\omega)] &= nN(0)(J^{cf})^2\left\{\tfrac{2}{3}S(S+1)\left[\frac{T}{T_N}\right]^{\nu}\tanh\left[\frac{\beta\omega}{2}\right]\right. \\ &\quad\times\delta(\omega-0^+) \\ &\quad\left.+S\left[1-\left[\frac{T}{T_N}\right]^{\nu}\right]\delta(\omega-\omega_s)\right\},\end{aligned} \tag{2.21a}$$

$$B^{zz}(\omega)=nN(0)(J^{cf})^2\tfrac{1}{3}S(S+1)\left[\frac{T}{T_N}\right]^{\nu}\tanh\left[\frac{\beta\omega}{2}\right]\times\delta(\omega-0^+)\,, \tag{2.21b}$$

where ω_s is the spin-wave frequency which is chosen to satisfy $\omega_s=k_BT_N$. Here, ν is the same exponent as in Eq. (2.20). For all $T>T_N$, the spectral weight functions are chosen to assume the values obtained from Eqs. (2.21) at $T=T_N$. Note that the elastic fluctuations vanish as T^{ν} when $T\to 0$, whereas the inelastic spin-fluctuation contributions increase as $1-(T/T_N)^{\nu}$ as T is lowered to 0. Above T_N, the spin fluctuations are all elastic, and thus behave as noninteracting magnetic impurities, having the appropriate value of τ_2. The spectral functions are related to the time-ordered propagators by the usual Lehmann representation. For example, for the phonons,

$$D(\omega_n-\omega_m)=\frac{1}{2\pi}\int_0^{\infty}d\omega\frac{B^{\rm ph}(\omega)}{\omega^2+(\omega_n-\omega_m)^2}\,. \tag{2.22}$$

In the Einstein model,

$$B^{\rm ph}(\omega)=\lambda\frac{\omega_E}{2}\delta(\omega-\omega_E)\,,$$

where the coupling constant $\lambda\equiv gN(0)$ and the phonon frequency ω_E are chosen to be 0.5 and ω_c, respectively.

It is important to note that the spin-fluctuation propagators are related to the temperature-dependent staggered magnetization $M(T)$ by the sum rule

$$nM^2(T)+\frac{1}{\beta(J^{cf})^2}\sum_n\sum_q\chi^{zz}(q;\nu_n)+\tfrac{1}{2}[\chi^{+-}(q;\nu_n)+\chi^{-+}(q;\nu_n)]=nS(S+1)\,, \tag{2.23}$$

where $M(T)\equiv\langle S^z\rangle_Q$ which is, in turn, proportional to the molecular field H_Q: $H_Q(T)=nJ^{cf}M(T)$. This sum rule indicates that the temperature dependence of the molecular field H_Q is dependent on that of χ. These parameters cannot be varied independently.

III. NUMERICAL RESULTS

A. Description of the technique

In the "strong-coupling" limit, when inelastic magnon and phonon processes are treated in full detail it is convenient to convert the equation for H_{c2} [Eq. (2.14)] to an eigenvalue problem following the method of Bergmann and Rainer.[23] Introducing an auxiliary parameter ρ, and considering discrete Matsubara frequencies, Eq. (2.14) can be rewritten as

$$\rho\Delta_n=\sum_{m=-\infty}^{\infty}K_{mn}\Delta_n\,, \tag{3.1a}$$

where

$$K_{mn}=2\pi\beta^{-1}\Gamma_{mn}-\delta_{mn}\left[\left[1-\frac{\tilde{H}_{Qm}^2}{\tilde{\omega}_m^2+\tilde{H}_{Qm}^2}\right]^{-1}\left[2(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}+\frac{e\hbar v_F^2}{c}H_{c2}/6(\tilde{\omega}_m^2+\tilde{H}_{Qm}^2)^{1/2}\right]-\hbar(\tau_1^{-1}-\tau_2^{-1})\right]\,, \tag{3.1b}$$

and

$$\Gamma_{mn} \equiv \lambda^{\rm ph}(m,n) - \tfrac{1}{2}[\chi^{+-}(m,n)+\chi^{-+}(m,n)] - \chi^{zz}(m,n) , \tag{3.1c}$$

with

$$\lambda^{\rm ph}(m,n) = \lambda\omega_E^2/[\omega_E^2+(\omega_n-\omega_m)^2] .$$

The linearized gap equation is satisfied whenever there exists a solution to Eq. (3.1c) with $\rho=0$. By obtaining these solutions as a function of T, one can thereby deduce the upper critical field.

The presence of AF order leads to a considerable complication in solving Eq. (3.1a). This is because the quantities $\tilde{\omega}_n$ and $\tilde{H}_{Qn}$ must also be determined self-consistently. In the strong-coupling case, to deal with the complexity of these equations we have made the additional assumption that the superconductor is sufficiently dirty so that

$$\hbar\tau_0^{-1} \gtrsim H_Q(0) \tag{3.2}$$

in all numerical work in this section. It should be noted, however, that the impurity concentration was not so high as to completely smear out the AF gap in the normal-state energy dispersion. Equation (3.2) is clearly a stronger condition than that necessary to ensure the validity of the quasiclassical approximation.

B. Numerical plots

In Fig. 2, experimental results are plotted for the upper critical field in some antiferromagnetic Chevrel[8] [Fig. 2(a)] and rhodium-boride[10] [Fig. 2(b)] compounds. In both figures, there is a pronounced structure at T_N. However, the $SmRh_4B_4$ system has an increased magnitude of slope at T_N, rather than a dip as seen in the Chevrel superconductors. Our numerical results will be qualitatively compared below with the data. It should be recalled that we have introduced errors into our calculations by neglecting paramagnetic effects. These effects are quite pronounced in $DyMo_6S_8$, and hence it is unlikely that our theory can treat this compound at any temperature. For the other AF superconductors the effects of Pauli paramagnetism can probably be neglected except for $T \approx T_N$.

Plotted in Fig. 3 are the results of a strong-coupling calculation of the H_{c2}-versus-T curve for $\tau_1^{-1}=2.0\omega_c$ and $\tau_2^{-1}=1.0\times10^{-4}\omega_c$ and $\nu=4$. Here, $H_Q(0)$ is varied while the spin-fluctuation coupling strength is held fixed. As in all of the examples, we have used a 1D model, although the effects of three dimensions can roughly be included by adding to each curve the usual[11] (nonmagnetic superconductor) contribution to H_{c2}. The effects of including this three-dimensional contribution are primarily to weaken the structure seen at $T \lesssim T_N$. It is clear from the figure that the larger the $H_Q(0)$, the more pronounced the dip at T_N in H_{c2}. For the smallest values of $H_Q(0)$ (curve 1) in Fig. 3 there is an increase in the magnitude of the slope below T_N, similar to what is seen in $SmRh_4B_4$. This increase arises because the onset of H_Q at T_N, according to

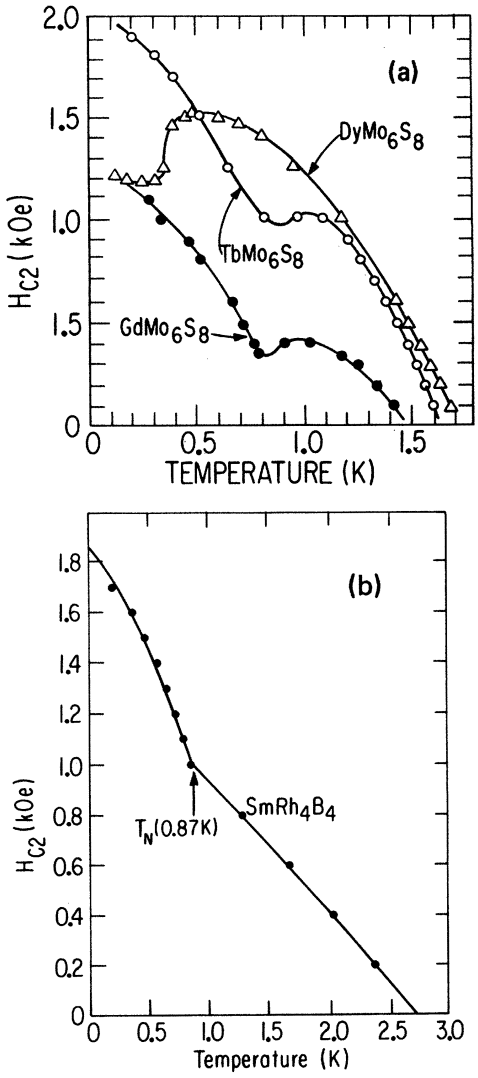

FIG. 2. Experimental results for H_{c2} in $SmRh_4B_4$, and in a series of Chevrel compounds. (a) is from Ref. 8 and (b) is from Ref. 10.

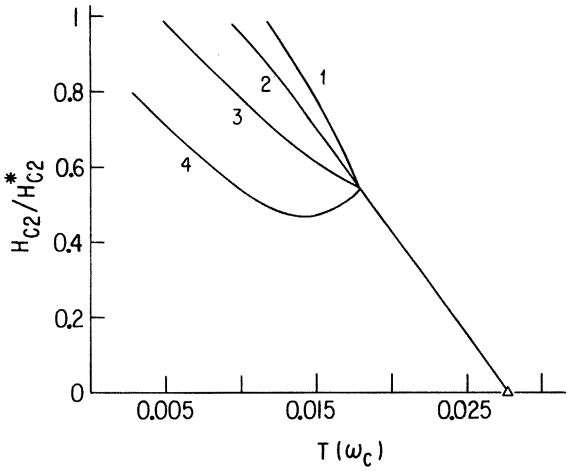

FIG. 3. Strong-coupling calculation of the T dependence (in units of ω_c) of H_{c2} and for $H_Q(0)$ equal to 0.05 (curve 1), 0.20 (curve 2), 0.30 (curve 3), and 0.04 (curve 4) in units of ω_c. All other parameters are held fixed.

the sum rule [Eq. (2.23)] must be compensated by a decrease in the spin-fluctuation scattering. For the parameters in curve 1 (Fig. 3) [for which $N(0)J^{cf}=3.6\times10^{-3}$], this decrease in pair breaking dominates over the increased pair breaking due to H_Q and thus causes H_{c2} to abruptly increase at T_N. In curve 3 of Fig. 3, when $N(0)J^{cf}=0.45\times10^{-3}$, the converse is true. The critical value of $N(0)J^{cf}$, at which there is a near cancellation of spin-fluctuation and molecular-field effects, is $\sim1.0\times10^{-3}$. Curve 2 in Fig. 3 corresponds to this case. It should be stressed that this critical value corresponds to a physically reasonable range of parameters in the ternary compounds.[10] This helps to explain why these systems exhibit both decreased and increased pair-breaking effects below T_N. In Ref. 3, decreased as well as increased pair breaking was observed but the former was found only in the unphysical case of $H_Q=0$.

Finally, in Fig. 4 we plot H_{c2} versus T for fixed $H_Q(0)=0.3\omega_c$ and $nN(0)(J^{cf})^2=1.8\times10^{-4}\omega_c$ and $\tau_2^{-1}=1\times10^{-4}\omega_c$ with the variable τ_1^{-1} in units of ω_c. The cleanest superconductor in Fig. 4 corresponds to curve 3, and the dirtiest corresponds to curve 1. As shown in Fig. 1, the effects of increasing the impurity concentration are generally to "screen" out the molecular field. Consequently, as τ_1^{-1} increases, H_{c2} also increases and the dip at T_N becomes less pronounced. As noted above the effects of decreasing spin-fluctuation scattering at T_N can then lead to an abrupt rise in H_{c2} at the Néel temperature. This was not seen in Fig. 1 where spin-fluctuation scattering was ignored. It should be noted that Figs. 3 and 4 are rather similar. Thus, changing $H_Q(0)$ or τ_1^{-1} can have analogous effects. However, the more relevant figure to compare with (future) experiments is Fig. 4, since τ_1 can be more readily varied than can the molecular field. As remarked in Sec. II, we have not explicitly included the effects of τ_1 on T_N, on the spin-fluctuation propagator χ, or on $H_Q(0)$. Since increasing τ_1^{-1} probably decreases $H_Q(0)$, this should act to enhance the "screening out" of the molecular field, as discussed above. The effects of decreasing T_N as the impurity concentration increases are primarily to lower the temperature at which the various structures at T_N, exhibited in the figures, will occur. The most serious approximation is probably the neglect of impurity effects on χ. Our approach is in the same spirit as the conventional treatment of dirty superconductors. However, the validity of this approximation cannot be established at present. Presumably, impurity effects in the bare molecular field H_Q and in χ will, at least partially, cancel.

A detailed test of our predictions might involve studies of pseudoternary systems in which small amounts of the transition element are substitutionally replaced in the Chevrel or rhodium-boride compounds. It should be noted that in high concentrations this disorder may change the nature of the magnetic and superconducting order and lead to effects beyond those included in our theoretical model. Replacement of the rare-earth ions by nonmagnetic elements should not be appropriate since this may introduce an effective τ_2 or spin-flip lifetime. The anomalous behavior that we are focusing on comes entirely from the τ_1 term.

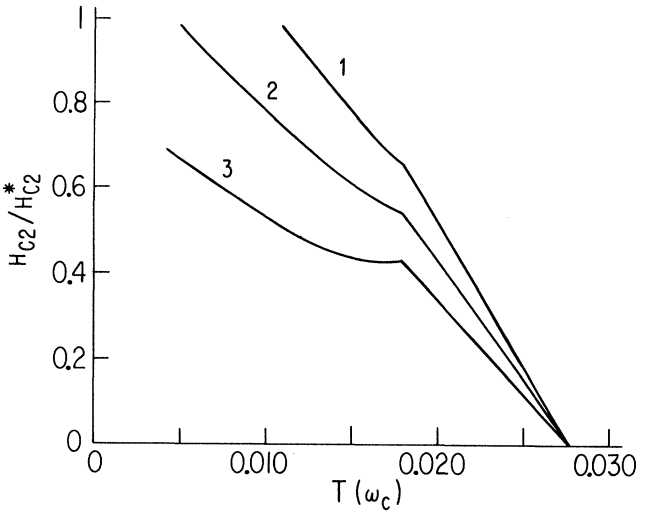

FIG. 4. Strong-coupling calculation of $H_{c2}(T)$ for various nonmagnetic impurity scattering times τ_1. Curves 1–3 correspond to $\tau_1=0.4$, 0.5, 0.666 (in units of ω_c), respectively.

In summary, qualitative comparison of Fig. 4 and (future) experiments can be used as an important test of the theory of $H_{c2}(T)$ presented here. Since a number of previous theories[2,3,5] have met with some success in explaining H_{c2} data, a means of discriminating between different theoretical mechanisms is essential. It should be pointed out that the effects of a nonspherical Fermi surface[4] and in some systems electromagnetic effects[2,24] may be important in addition to the competition between molecular-field and spin-fluctuation scattering that we have invoked here. However, the assumption of magnetic quasiparticle pairing[5,6] appears to violate self-consistency at several levels.[25] Our predictions for the effects of nonmagnetic impurities on H_{c2} may help sort out the nature of the interactions between magnetic and superconducting order. Furthermore, it should be recognized that H_{c2} measurements provide a way of establishing the degree to which nonmagnetic impurities are pair breaking in AF superconductors. This effect, which is rather unique to the AF superconductors, may be quite dramatic.

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