Theory of antiferromagnetic superconductors

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(Received 29 January 1980)

In this paper a theory is presented of antiferromagnetic superconductors in which a spin-density wave (SDW) ordering with a wave vector $\vec{Q}$ may coexist with superconductivity. The effect of the antiferromagnetic molecular field $h_Q(T)$ on the Cooper pairing is studied, and it is shown that, below the magnetic transition temperature T_N, the Bardeen-Cooper-Schrieffer coupling parameter is reduced by a factor $[1-\text{const}|h_Q(T)|/\epsilon_F]$ due to the formation of energy gaps of SDW on the Fermi surface along $\vec{Q}$ and this reduction can explain the anomaly in the upper critical field H_{c2} just below T_N as observed in $R\text{Mo}_6\text{S}_8$ (R = Gd, Tb, and Dy). Taking account of both the spin-orbit scattering and spin-fluctuation effect near T_N in addition to the effect of $h_Q(T)$, a theoretical calculation of the superconducting transition temperataure T_c and $H_{c2}(T)$ is performed. Detailed quantitative comparisons between theory and experiments on $H_{c2}(T)$ are made with fairly good accord for the above three compounds. Some speculation is devoted to the remaining discrepancies between theory and experiments and certain phenomena not yet explained by the theory.

I. INTRODUCTION

Since new superconducting materials in which magnetic moments are built in to form a regular lattice have been synthesized, new light is shed on the "old problem" of the coexistence of magnetism and superconductivity.[1] It is evident from experiments on the rare-earth ternary compounds (HoMo_6S_8 and ErRh_4B_4) that the onset of ferromagnetism at low temperatures simply destroys superconductivity, and that these two types of long-range order cannot coexist in general except at a very narrow temperature region for which a definite conclusion is not yet obtained. On the other hand, there is no *a priori* reason to exclude the coexistence of antiferromagnetism and superconductivity, because periodicity of antiferromagnetic ordering (usually a few angstroms) is so short compared with the superconducting coherent length (of the order of a few hundred angstroms). Then spin fluctuations are averaged out and are ineffective in destroying the superconducting state. Historically, several authors[2] predict the coexistence of antiferromagnetism and superconductivity. In fact such coexistence has been found recently in several ternary compounds with rare-earth atoms: the Chevrel phase[3] $R\text{Mo}_6\text{S}_8$ (R = Gd, Tb, and Dy) and the rhodium-boride phase[4] SmRh_4B_4.

In the former compounds, neutron experiments[5] have been done to identify their magnetic structures, which are sinusoidal spin-density-wave states along the crystal c axis commensurate with the underlying crystal lattice.

A basic picture of the electronic origin to describe such ternary rare-earth compounds is established and widely accepted now; that is, a characteristic of these compounds is to have partially filled localized $4f$ electrons which are responsible for magnetism. These local moments interact very weakly with conduction electrons mainly coming from d bands via exchange interaction I. Moreover, one of the dominant mechanisms which mediate between local moments is the indirect-exchange-RKKY (Ruderman-Kittel-Kasuya-Yosida) interaction via the conduction electrons[6] which play an important role in superconductivity.

In a series of papers[7,8] one of the authors has investigated the spin-fluctuation effect of both ferromagnetic[7] and antiferromagnetic cases[8] on superconductivity to generalize the Abrikosov-Gorkov theory of paramagnetic impurities to correlated spin systems. This spin-fluctuation theory succeeds in explaining several aspects of coexistence problem: It gives a correct explanation for the bell-shaped upper critical field (H_{c2}) in the ferromagnetic case ErRh_4B_4 and for systematic change in the superconducting transition temperatures (T_c) for $R\text{Mo}_6\text{S}_8$ (R being heavy rare-earth atoms). However, this theory fails to explain the anomaly in H_{c2} near the Néel temperature (T_N), which is commonly observed in the antiferromagnetic superconductors of the Chevrel phase.

In this paper we shall present a theory of antiferromagnetic superconductors that suppliments our previous theory taking into account the effects of the antiferromagnetic molecular field exerted by aligned local moments in addition to the spin-fluctuation effect, because the antiferromagnetic energy gap formed on the Fermi surface perturbs the superconducting state below T_N. This is a similar situation to that dis-

cussed by Fulde and Ferrell[9] in which superconducting pairings are modified by the ferromagnetic molecular field. As we have discussed previously,[10] if the Fermi surface is highly anisotropic or one-dimensional-like (in other words if Fermi-surface nesting is large enough), then we have demonstrated that the superconducting state is heavily perturbed by the presence of anitiferromagnetic energy gap. In this paper we only consider an ordinary three-dimensional Fermi-surface model to clarify the situation. We also restrict ourself to the magnetic order of the spin-density-wave type characterized by a single wave vector $\vec{Q}$. This is the case in the Chevrel phase compounds.

Organization of the paper is as follows: The next section discusses stability of a superconducting state under a periodic antiferromagnetic molecular field and introduces a simple band model to describe the perturbed superconducting state by antiferromagnetism. In Sec. III we calculate the temperature dependence of the order parameter and the upper critical field. In Sec. IV, in addition to the simplified band model, we take into account the spin-fluctuation effect and analyze the experimental data of H_{c2} on $R\,Mo_6S_8$ (R = Gd, Tb, and Dy). The final section will be devoted to discussion and conclusion.

II. STABILITY OF SUPERCONDUCTING STATE AND SIMPLIFIED BAND MODEL

We take the following "s-f" exchange Hamiltonian:

$$H=\sum \xi_k C^{\dagger}_{k\sigma}C_{k\sigma} - \frac{I}{2N}(g_J-1)\sum \vec{J}_i\cdot\vec{\sigma}_{\mu\nu}C^{\dagger}_{k\mu}C_{k'\nu}e^{i(k-k')R_i} , \tag{2.1}$$

where the conduction electrons interact with the $4f$ moment J_i localized at the lattice site i of rare-earth ion and g_J is the Landé g factor. Below T_N the sublattice magnetization J_Q of the antiferromagnetic state becomes nonvanishing where J_Q is the Qth component of the Fourier transform of J_i and $\vec{Q}$ is the wave vector characterizing the antiferromagnetic state. Therefore the $4f$ local moment system exerts a spatially periodic antiferromagnetic molecular field on the conduction-electron system. The above Hamiltonian reduces

$$H=\sum_{k\sigma}\xi_k C^{\dagger}_{k\sigma}C_{k\sigma}-\sum_{k\sigma}(h_Q\sigma C^{\dagger}_{k\sigma}C_{k+Q\sigma}+\text{H.c.}) \tag{2.2}$$

below T_N, where

$$|h_Q|^2=\frac{I^2}{(2N)^2}(g_J-1)^2|J_Q|^2 . \tag{2.3}$$

We now consider instability of a superconducting state under a spatially periodic molecular field h_Q to examine what kind of pairing states are the most stable. The stability may be investigated by the following linearized gap equation[11] for a spatially varying order parameter $\Delta(r)$:

$$\Delta(r)=gT\sum_{\omega_n}\int K(r,r',\omega_n)\Delta(r')dr' , \tag{2.4}$$

$$K(r,r',\omega_n)=G^{\uparrow}(r,r',\omega_n)G^{\downarrow}(r,r',-\omega_n) , \tag{2.5}$$

where $G^{\sigma}(r,r',\omega_n)$ is the thermal Green's function of the normal state under the antiferromagnetic molecular field. The Fourier transformation of the integral kernel $K(r,r',\omega_n)$ is given by

$$K(p,-p',\omega_n)=\sum_{k_i}G^{\uparrow}(k_1,k_2,\omega_n)G^{\downarrow}(k_3,k_4,-\omega_n)\delta(k_1+k_3-p)\delta(k_2+k_4-p') . \tag{2.6}$$

Assuming $Q=2k_F$ (k_F is the Fermi wave number) to simplify the following argument,[12] let us evaluate $K(0,0,\omega_n)$, that is,

$$\begin{aligned}K(0,0,\omega_n)&=\sum_{kk'}G^{\uparrow}(k,k',\omega_n)G^{\downarrow}(-k,-k',-\omega_n)\\&=\sum_{k}[G^{\uparrow}(k,k,\omega_n)G^{\downarrow}(-k,-k,-\omega_n)+G^{\uparrow}(k,k-Q,\omega_n)G^{\downarrow}(-k,-k+Q,-\omega_n)\\&\quad+G^{\uparrow}(k,k+Q,\omega_n)G^{\downarrow}(-k,-k-Q,-\omega_n)] ,\end{aligned} \tag{2.7}$$

where

$$G^{\sigma}(k,k,\omega_n)=\frac{i\omega_n-\xi_{k-Q}}{(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2} , \quad G^{\sigma}(k,k-Q,\omega_n)=\frac{-\sigma h_Q}{(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2} . \tag{2.8}$$

The basic assumptions we now make are (1) the spherical Fermi surface of the three dimensions and (2) $|h_Q| \ll \epsilon_F$ where ϵ_F is the Fermi energy. From Eq. (A8) in the Appendix we can evaluate $K(0,0,\omega_n)$ as

$$K(0,0,\omega_n)=\frac{\pi N(0)}{|\omega_n|}\left(1-\frac{\pi|h_Q|}{4\epsilon_F}\right) , \tag{2.9}$$

where $N(0)$ is the density of states at the Fermi energy. In a similar way from Eq. (A10) the off-diagonal element $K(Q,0,\omega_n)$ is given by

$$K(Q,0,\omega_n)=\sum_k [G^{\uparrow}(k+Q,k+Q,\omega_n)G^{\downarrow}(-k+Q,-k-Q,-\omega_n) + G^{\uparrow}(k+Q,k-Q,\omega_n)G^{\downarrow}(-k+Q,-k+Q,-\omega_n)] = N(0)/16\epsilon_F . \tag{2.10}$$

Then it turns out that since the off-diagonal elements such as $K(Q,0,\omega_n)$ are negligibly small $[\cong O(T/\epsilon_F)]$ compared with the diagonal element $K(0,0,\omega_n)$, the simple Bardeen-Cooper-Schrieffer (BCS) state characterized by a spatially uniform order parameter is most stable. Therefore, it readily yields to the gap equation of the BCS type:

$$\Delta = gN(0)\left(1-\frac{\pi|h_Q|}{4\epsilon_F}\right)\pi T\sum_{\omega_n}\frac{\Delta}{|\omega_n|} . \tag{2.11}$$

This shows that the effective attractive interaction $\tilde{g}N(0)$, or equivalently the density of states at the Fermi energy, is diminished by the periodic molecular field, that is,

$$\tilde{g}N(0)=gN(0)[1-\alpha m(T)] , \tag{2.12}$$

$$\alpha=\tfrac{1}{4}\pi(g_J-1)[J(J+1)]^{1/2}\frac{|I|/\epsilon_F}{gN(0)} , \tag{2.13}$$

$$m(T)=\frac{J_Q(T)}{J_Q(0)} , \tag{2.14}$$

where $m(T)$ is the normalized sublattice magnetization of the antiferromagnetic state. The physical meaning of Eq. (2.12) is that the particular portions of the pairing states on the Fermi surface, which are brought together by the translation Q, are mixed strongly to yield antiferromagnetic energy gaps. These states on the Fermi surface are unable to participate in forming a superconducting state antiferromagnetic ordering takes place. So far the spherical Fermi-surface model has been used. A closed examination of the above derivation together with the Appendix B where a simplified and intuitive derivation of Eq. (2.12) is given reveals that the detailed shape of the Fermi surface delicately affects evaluation of α and changes its numerical factor. Because detailed information on the band structure of the ternary compounds we are now considering is not available at present, we regard α as an adjustable parameter. However, we should keep in mind that α is roughly of an order of 0.1, which is estimated by substituting typical values for the parameters $I=112.2$ K (we will derive this value later), $\epsilon_F \cong 5000$ K (from the band calculation in similar compounds[13]), $(g_J-1)\times[J(J+1)]^{1/2}\cong 4$ (the free-ion value for Gd), and $gN(0)\cong 0.2$ into Eq. (2.13).

III. ORDER PARAMETER AND THE UPPER CRITICAL FIELD

In this section we determine the temperature dependence of the order parameter $\Delta(T)$ and the upper critical field H_{c2} on the basis of the simplified band model derived in the previous section. The basic equation for $\Delta(T)$ is given by the BCS theory in which the effective attractive interaction is simply replaced by the temperature-dependent one $\tilde{g}N(0)$ which is a function of $m(T)$. The self-consistent equation of $\Delta(T)$ is given by

$$\Delta(T)=\tilde{g}N(0)\sum_{\omega_n}\frac{\Delta(T)}{\omega_n^2+\Delta^2(T)} , \tag{3.1}$$

where $\tilde{g}N(0)$ is determined by Eq. (2.12). Near $T=0$ the order parameter reduces to the following BCS form:

$$\frac{\Delta(T)}{\Delta(0)}=1-\left(\frac{2\pi T}{\Delta(0)}\right)^{1/2}\left(1-\frac{T}{8\Delta(0)}\right)e^{-\Delta(0)/T} , \tag{3.2}$$

$$\Delta(0)=\Delta_0 e^{-\alpha} , \tag{3.3}$$

where $\Delta_0(T_{c0})$ is the order parameter (transition temperature) of the pure BCS superconductor without local moments. Between T_{c0} and T_N (we assume $T_{c0}>T_N$) the order parameter is nothing but that of the BCS theory. Since below T_N the magnetization $m(T)$ becomes nonvanishing, $\tilde{g}N(0)$ is weakened which results in a sudden drop of $\Delta(T)$ immediate below T_N. As $m(T)$ saturates at lower temperatures, $\Delta(T)$ gradually recovers its value with increasing the superconducting condensation energy. As we can see from the functional form of Eq. (3.3), the supercon-

ducting state is never destroyed by antiferromagnetic ordering as long as our approximation ($|h_Q| \ll \epsilon_F$) is valid. If the spin-fluctuation effect is taken into account, this is not the case, as we will see later.

If α is large enough ($\alpha \geq \alpha_{cr} \cong 0.8$ for $J=\frac{1}{2}$), on decreasing temperature superconductivity ceases to exist by the onset of antiferromagnetism to reentrant to the normal state at $T=T_u$ and then appears once again at a lower temperature T_l. The order parameter $\Delta(T)$ behaves near $T=T_u$ or T_l for $\alpha \geq \alpha_{cr} \cong 0.8$

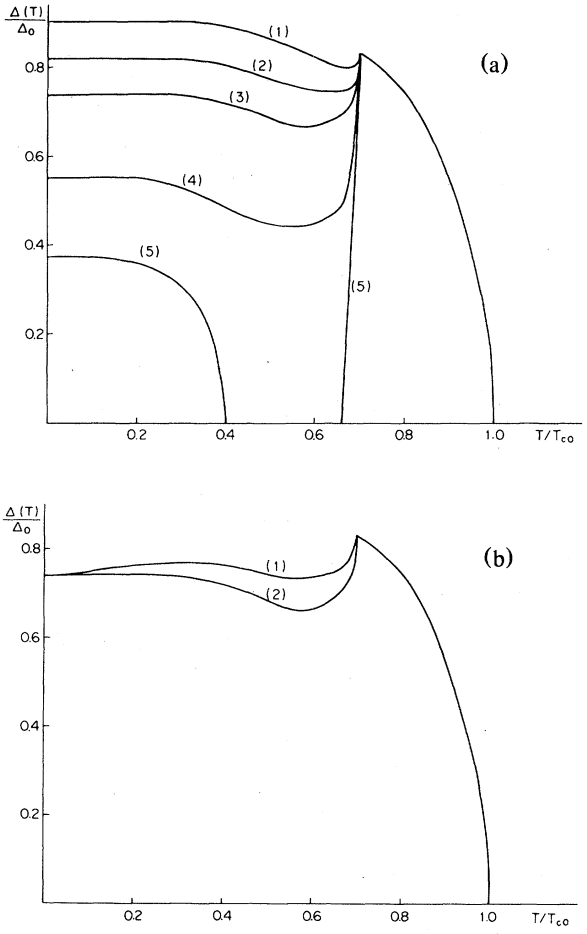

FIG. 1. (a) Temperature dependence of the order parameter $\Delta(T)$ for $J=\frac{1}{2}$ and $T_N/T_{c0}=0.7$: Curve (1) $\alpha=0.1$, curve (2) 0.2, curve (3) 0.3, curve (4) 0.6, and curve (5) 1.0. Curve (5) shows the interrupted superconductivity in which the normal state appears in $0.40 \leq T/T_{c0} \leq 0.62$. (b) Temperature dependence of the order parameter $\Delta(T)$ for $\alpha=0.3$, $T_N/T_{c0}=0.7$: Curve (1) $J=\infty$ and curve (2) $J=\frac{1}{2}$. Note that the total angular momentum J affects the shapes of $\Delta(T)$ near T_N.

($J=\frac{1}{2}$ case) as follows:

$$\frac{\Delta(T)}{\Delta_0} = e^{\gamma}\left(\frac{8}{7\zeta(3)}\right)^{1/2} e^{-\alpha m(T_i)} \times \left|1+\alpha T_i\left(\frac{dm}{dT}\right)_{T_i}\right|^{1/2}\left|1-\frac{T}{T_i}\right|^{1/2}, \quad (3.4)$$

$$i = u \text{ or } l \ ,$$

where γ is the Euler constant and $\zeta(3)$ is the zeta function. We will further discuss this "interrupted superconductivity" later. In Fig. 1 we depict several numerical examples of $\Delta(T)$ where the normalized sublattice magnetization $m(T)$ is assumed to be given by the Brillouin function. Note that the total angular momentum J delicately influences on the shape of $\Delta(T)$ near T_N. Sharp rise of $m(T)$ near T_N causes a pronounced drop in $\Delta(T)$ at T_N. We compare two cases of $J=\frac{1}{2}$ and ∞ in Fig. 1(b).

It should be emphasized that even a fairly small value α drastically suppresses superconductivity; for instance, $\Delta(0)$ is half of Δ_0 when $\alpha=0.1[gN(0)=0.2]$. This means that destruction of a small area on the Fermi surface due to the onset of antiferromagnetism results in a drastic change of the superconducting state. This becomes important later when interpreting experimental data.

The equation for the upper critical field[14] H_{c2} is given by the standard formula[15] except that $\tilde{g}N(0)$ is replaced according to Eq. (2.12); that is, in the dirty limit (the diffusion constant D is small) it is given by

$$\alpha m(T) + \ln\frac{T}{T_{c0}} + \psi\left(\frac{1}{2}+\frac{DeB}{2\pi T}\right) - \psi\left(\tfrac{1}{2}\right) = 0 \ , \quad (3.5)$$

where we have neglected change in $m(T)$ due to applied magnetic field. A detailed analysis of this equation will be given in the next section.

IV. SPIN-FLUCTUATION EFFECT AND DATA ANALYSIS OF H_{c2}

In the previous section we have investigated several characteristics of the superconducting state under the periodic antiferromagnetic molecular field. In order to check our idea on the truncated Fermi-surface model, and to analyze experimental data of H_{c2} on the series of the rare-earth Chevrel-phase compounds, we must take into account spin-fluctuation effect[8] both above and below T_N, which gives rise to a finite lifetime in conduction-electron motion. This effect gives a satisfactory explanation to some aspects of experiments. Firstly, we briefly summarize our previous theory[8] of the spin-fluctuation effect.

We treat the exchange interaction I within the Born approximation because $IN(0)$ is very small. The relaxation time $\tau(T)$ of the conduction-electron system

is given by

$$\frac{1}{\tau(T)} = 2\pi N(0)(\tfrac{1}{2}I)^2(g_J-1)^2\frac{1}{4\pi}\int d\Omega_q\, S_q(T) \ , \tag{4.1}$$

$$S_q(T) = \langle \vec{J}_q \cdot \vec{J}_{-q} \rangle \ , \tag{4.2}$$

where $S_q(T)$ is the correlation function between local moments. We assume that the most important spin-fluctuation effect on the superconducting state comes short wavelengths, $q=0$. Since the superconducting coherence length is so large, and small-wavelength fluctuations are averaged out on that scale, then long-wavelength fluctuations are effective in suppressing the superconducting state. We adopt a mean-field Curie-Weiss-type correlation function for $S_0(T)$: When $T \geq T_N$,

$$S_0(T) = \frac{J(J+1)T}{T+T_N} \ , \tag{4.3}$$

when $T < T_N$,

$$S_0(T) = \tfrac{1}{3}[S_{\parallel}(T) + 2S_{\perp}(T)] \ , \tag{4.4}$$

$$S_{\parallel}(T) = \frac{J(J+1)}{2}\frac{T}{T_N} \ , \tag{4.5}$$

$$S_{\perp}(T) = \frac{3TJ^2B_J'(2zJ|\mathcal{J}|J_Q/T)}{T+2z|\mathcal{J}|J^2B_J'(2zJ|\mathcal{J}|J_Q/T)} \ . \tag{4.6}$$

We have introduced the Brillouin function $B_J(x)$, its first derivative $B_J'(x)$ and the number of the nearest neighbors of a magnetic ion, z. The thermal average J_Q is determined by $J_Q = JB_J(2zJ|\mathcal{J}|J_Q/T)$, where $|\mathcal{J}|$ is the exchange integral between $4f$ moments, and gives the Néel temperature $T_N = \frac{2}{3}zJ(J+1)|\mathcal{J}|$. Since conduction electrons moving through a sample frequently change direction, the fluctuation field they feel should be averaged over all directions. The relaxation time thus reduces to

$$1/\tau(T) = 2\pi N(0)(\tfrac{1}{2}I)^2(g_J-1)^2 S_0(T) \ . \tag{4.7}$$

A. Transition temperature

The depression of the superconducting transition temperature T_c is determined by

$$-\ln\frac{T_c}{T_{c0}} = \psi(\tfrac{1}{2}+\rho(T_c)) - \psi(\tfrac{1}{2}) \tag{4.8}$$

with the pair-breaking parameter $\rho(T_c) \equiv 1/[2\pi \times T_c\tau(T_c)]$ and where T_{c0} is the hypothetical transition temperature with $I=0$. We now consider the series of the heavy rare-earth antiferromagnetic Chevrel compounds RMo_6S_6 (R = Gd, Tb, and Dy). Using the same procedure as before,[8] that is, assuming (1) the free-ion values of each rare-earth atom for J and g_J, (2) the hypothetical transition temperature T_{c0} to be determined by the interpolation of the two transition temperatures of the end members $LaMo_6S_8$ and $LuMo_6S_8$ in the Periodic Table as done by Ishikawa and Muller,[16] and (3) I being held constant for three compounds, then we evaluate T_c as shown in Table I. Here we have chosen a slightly larger value $I = 112.2$ K than before ($I = 104$ K).

B. Upper critical field

It is easy to generalize the calculation for H_{c2} in the previous section to take into account the spin-fluctuation effect. Combining the discussions in the pre-

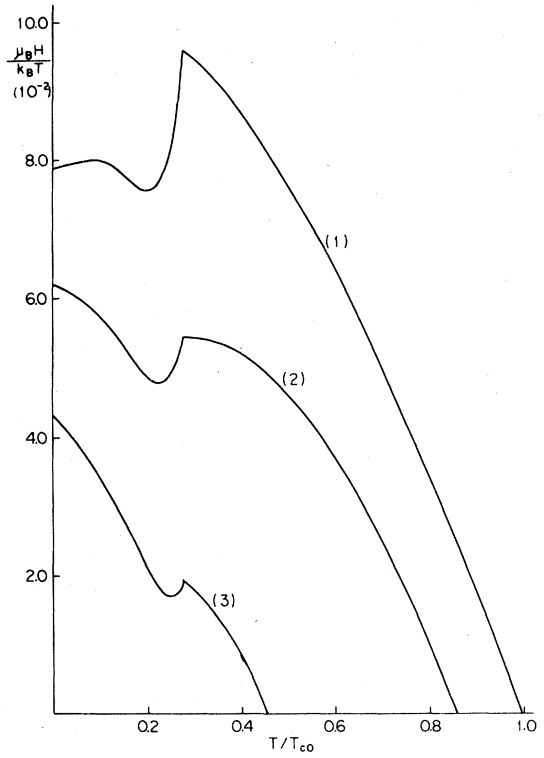

FIG. 2. Upper critical field H_{c2} as a function of temperature for several values of I/T_{c0}: Curve (1) $I/T_{c0}=0$, curve (2) 15, and curve (3) 31.16. The following parameters are used $De/2\pi\mu_B = 1.0$, $1/6\pi\tau_{so}T_{c0} = 350$, $T_N/T_{c0} = 0.28$, $J=6$, $g_J = \frac{3}{2}$, and $\alpha = 0.4$. Curve (1) corresponds to the case without the spin-fluctuation effect.

vious paper[8] (Sec. 4) and in the Sec. III, we readily obtain the equation for H_{c2} in the dirty limit as follows:

$$\alpha m(T)+\ln\frac{T}{T_{c0}}+\frac{1}{2}\left[\left(1+\frac{b}{(b^2-h^2)^{1/2}}\right)\psi\left(\frac{1}{2}+\rho_-\right)+\left(1-\frac{b}{(b^2-h^2)^{1/2}}\right)\psi\left(\frac{1}{2}+\rho_+\right)\right]-\psi\left(\frac{1}{2}\right)=0 \ , \tag{4.9}$$

where

$$\rho_\pm=\frac{1}{2\pi T}\left[\frac{1}{\tau(T)}+\frac{1}{\tau_{so}}+DeB\pm(b^2-h^2)^{1/2}\right] \ , \tag{4.10}$$

$$b=\frac{1}{\tau_{so}}=\frac{1}{2}n_{so}N(0)\int d\Omega\,|v_{so}|^2\sin^2\theta \ , \tag{4.11}$$

$$h=g_J\mu_B IS_0(T)H/T+\mu_B B \ . \tag{4.12}$$

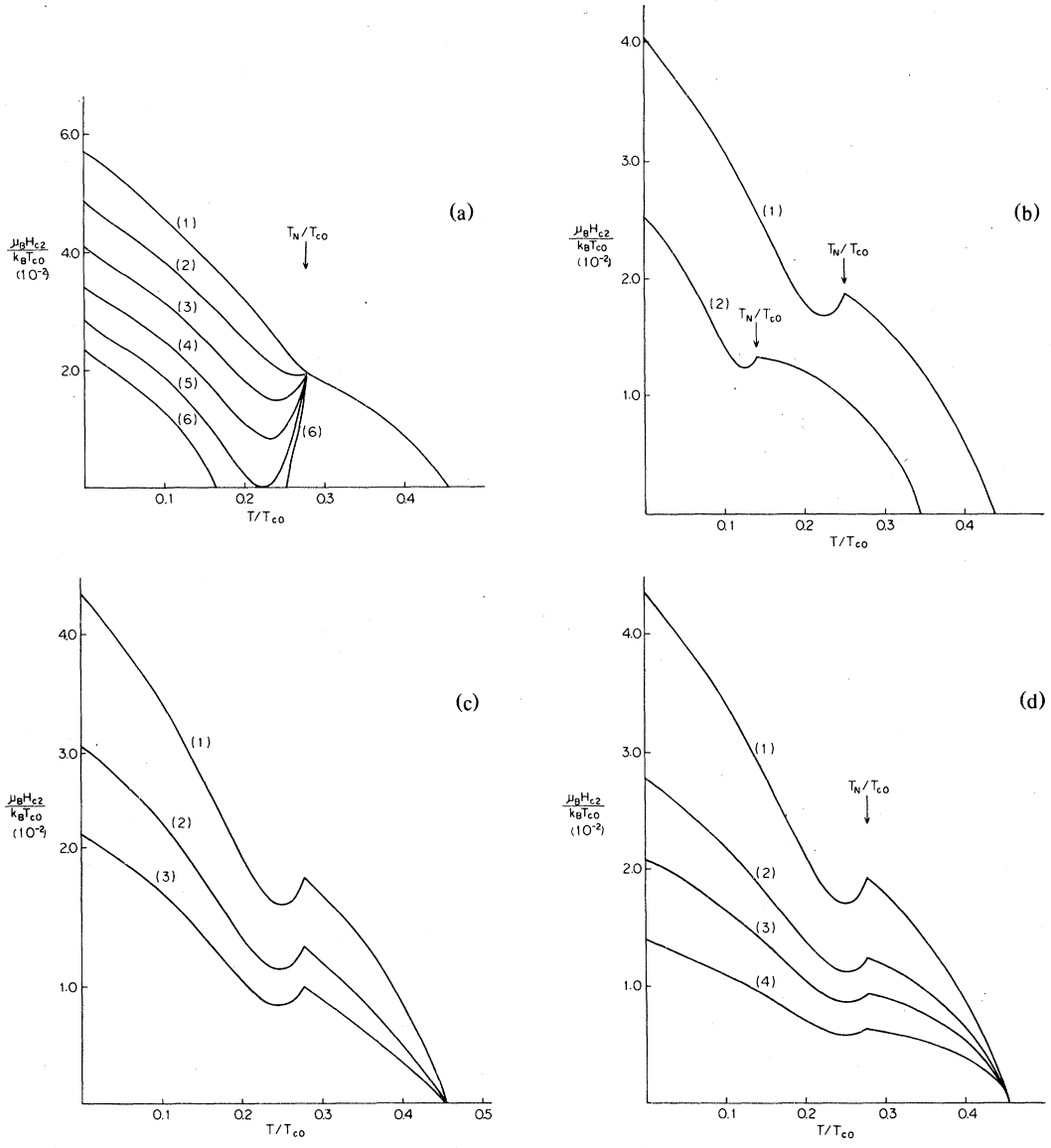

FIG. 3. Upper critical field H_{c2} as a function of temperature. The following parameters are used $De/2\pi\mu_B=1.0$, $1/6\pi\tau_{so}T_{c0}=350$, $J=6$, $g_J=\frac{3}{2}$, $I/T_{c0}=31.16$, and $\alpha=0.4$, unless stated otherwise. (a) Curve (1) $\alpha=0.0$, curve (2) 0.25, curve (3) 0.50, curve (4) 0.75, curve (5) 1.00, and curve (6) 1.25. Curve (6) corresponds to the intervened superconductivity in which the normal state appears between $0.16\leqslant T/T_{c0}\leqslant 0.25$. (b) Curve (1) $T_N T_{c0}=0.25$ and curve (2) 0.14. (c) Curve (1) $De/2\pi\mu_B=1.0$, curve (2) 2.0, and curve (3) 3.0. (d) Curve (1) $1/6\pi\tau_{so}T_{c0}=350$, curve (2) 100, curve (3) 50, and curve (4) 20.

The effect of the spin-orbit scattering b and of the induced molecular field h by applied field have been included.

In Figs. 2 and 3 we show typical numerical results for H_{c2}. Curve (1) in Fig. 2 corresponds to the calculation in the Sec. III where the spin fluctuation is neglected. In Fig. 3(a) curve (1) for $\alpha = 0.0$ corresponds to the calculation in the previous paper,[8] and curve (6) for $\alpha = 1.25$ shows that superconductivity in interrupted by the normal state in the intermediate temperature region.

C. Analysis of the experimental data for H_{c2}

We now come to data analysis of the three rare-earth Chevrel compounds[3] $R\mathrm{Mo_6S_8}$ (R = Gd, Tb, and Dy). We use the following procedure to fit the data: (1) We use the same set of parameters (J, g_J, T_{c0}, and T_N) as were adopted in Table I for each compound keeping $I (= 112.2\ \mathrm{K})$ and $N(0) (= 0.2\ \mathrm{eV}^{-1})$ constant. (2) The remaining parameters D, b, and α are adjusted to reproduce the data.

The results are shown in Figs. 4–6. The overall feature of H_{c2} for all three compounds is reproduced for the experimental data fairly well; that is, we can successfully explain the abrupt changes of H_{c2} below T_N which are a common characteristic of the data in all three Chevrel-phase compounds of antiferromagnetic superductors. However, in $\mathrm{DyMo_6S_8}$ (Fig. 6) the theoretical curve (dotted curve) deviates from the data at low temperatures even if we choose an extremely short spin-orbit scattering rate ($1/6\pi\tau_{so}T_{c0} = 5000$). As shown in the same figure we have depicted the other theoretical curve (solid curve) which corresponds to the theory without the spin-fluctuation effect ($I = 0$). This improves the fit at low temperatures considerably. It might be possible in $\mathrm{DyMo_6S_8}$ to neglect the spin-fluctuation effect which is suppressed by external applied fields at low temperatures because applied fields are relatively strong in this temperature region.

TABLE I. Superconducting transition temperatures for $R\mathrm{Mo_6S_8}$ compounds [$I = 112.2$ K, $N(0) = 0.2/\mathrm{eV}$].

	T_{c0}	T_N	T_c (expt)	T_c (theor)
Gd	4.50	0.97	1.2 ~ 1.5	1.49
Tb	3.60	1.00	1.4 ~ 2.05	1.64
Dy	3.40	0.40	1.7 ~ 2.15	1.69

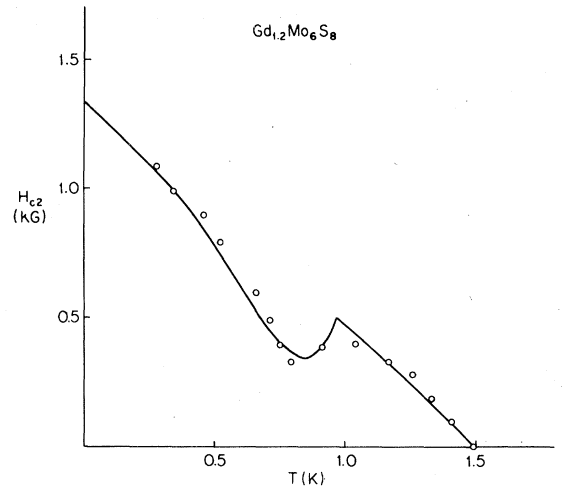

FIG. 4. $\mathrm{GdMo_6S_8}$. The following parameters are used $I = 112.2$ K, $T_N = 0.97$ K, $T_{c0} = 4.5$ K, $De/2\pi\mu_B = 3.0$, $1/6\pi\tau_{so}T_{c0} = 80$, and $\alpha = 0.5$. The free-ion values are used for $J = \frac{7}{2}$ and $g_J = 2$ (Gd).

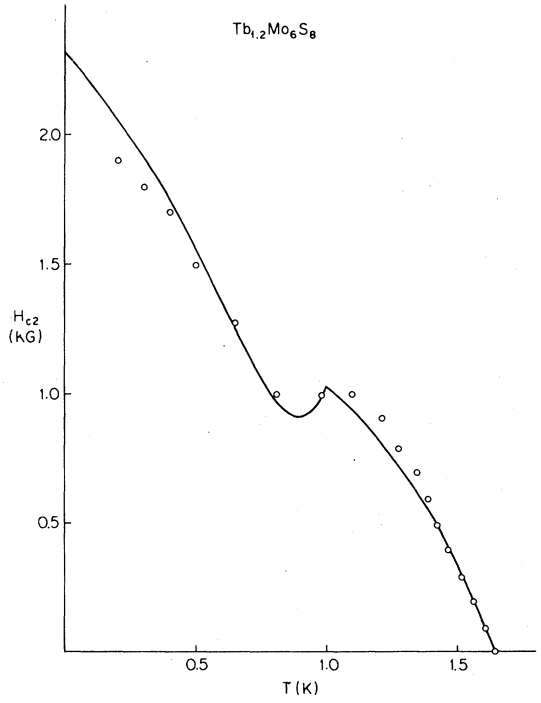

FIG. 5. $\mathrm{TbMo_6S_8}$. The following parameters are used $I = 112.2$ K, $T_N = 1.0$ K, $T_{c0} = 3.6$ K, $De/2\pi\mu_B = 1.0$, $1/6\pi\tau_{so}T_{c0} = 350$, and $\alpha = 0.4$. The free-ion values are used for $J = 6$ and $g_J = \frac{3}{2}$ (Tb).

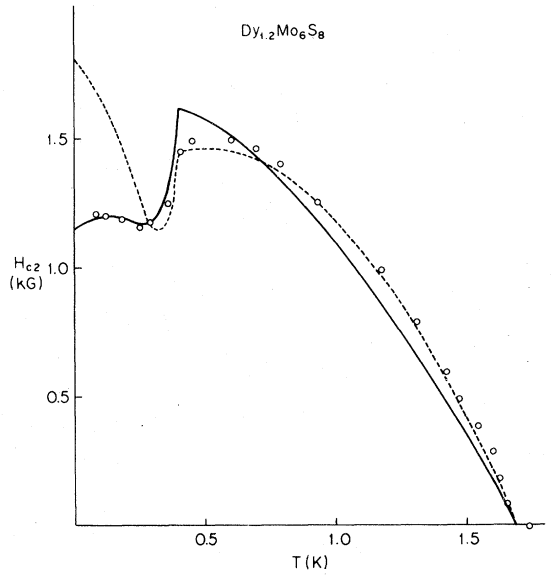

FIG. 6. $DyMo_6S_8$. $T_N = 0.4$ is assumed and the free-ion values are used for $J = \frac{15}{2}$ and $g_J = \frac{4}{3}$ (Dy) for both curves. The dotted curve with the spin-fluctuation effect: The other parameters are $I = 112.2$ K, $T_{c0} = 3.40$ K, $De/2\pi\mu_B = 1.0$, $1/6\pi\tau_{so}T_{c0} = 5000$, and $\alpha = 0.6$. The solid curve without the spin-fluctuation effect: The other parameters are $I = 0$ K, $T_{c0} = 1.67$ K, $De/2\pi\mu_B = 1.5$, $1/6\pi\tau_{so}T_{c0} = 100$, and $\alpha = 0.5$

V. DISCUSSION AND CONCLUSION

We have developed a theory of superconductivity which coexists with antiferromagnetic ordering and have demonstrated that the superconducting state can be well described by a model in which the effective interaction between conduction electrons is modified and reduced by the formation of the antiferromagnetic energy gap. In addition to this we have taken into account the scattering effects or spin-fluctuation effects, which lead to finite lifetime of conduction electrons, due to localized $4f$ moments in order to analyze the experimental data of T_c and H_{c2} on the rare-earth Chevrel-phase compounds.

The upper critical-field data of the antiferromagnetic superconductors RMo_6S_8 (R = Gd and Tb) are well described by the present model, by assuming the free-ion values for J and g_J and a constant I (112.2 K) and by adjusting the three parameters (the diffusion constant, the spin-orbit scattering rate, and α). On the other hand, H_{c2} of $DyMo_6S_8$ does not seem to follow the present model. Rather, it fits well with the theory neglecting the spin-fluctuation effect. It is interesting to note that H_{c2} of $SmRh_4B_4$, which is another ternary compound of the antiferromagnetic superconductor, can be explained quite satisfactorily by the present theory with $\alpha = 0$.[4]

In the context of the present theory we may classify these four antiferromagnetic superconductors into three groups: (1) The fluctuation dominant superconductor, $SmRh_4B_4$. It might be possible that for band-structural reasons the wave vector Q is greater or smaller than $2k_F$; therefore, the antiferromagnetic order hardly affects on the superconducting state. This means α is very small in the present model. It is highly desirable to determine the antiferromagnetic structure by neutron experiments, which has not been done yet. (2) The nonfluctuation superconductor, $DyMo_6S_8$. In this case the high applied magnetic field suppresses the spin fluctuation. The formation of the antiferromagnetic energy gap on the Fermi surface is the most important for this superconductor. (3) The intermediate-type superconductors, $GdMo_6S_8$ and $TbMo_6S_8$. We must take into account both effects, spin fluctuation and partial destruction of the Fermi surface due to the antiferromagnetic energy gap.

Needless to say, it is possible to interprete the results in the previous section in a different way, partly because in $DyMo_6S_8$ the spin-flipping applied magnetic field[16] which destroys the antiferromagnetic state is so low ($\cong 1.2$ kG) compared with the relatively high H_{c2} at low temperatures. Thus the anomalous behavior of H_{c2} in $DyMo_6S_8$ at low temperatures in which our theory markedly deviates from the data might be attributed to this magnetic-phase change under applied magnetic fields. This point remains unsettled.

As we have mentioned before if we substitute $I = 112.2$ K, $N(0) = 0.2$ eV^{-1}, and $gN(0) = 0.2$ into Eq. (2.13) we can estimate $\alpha \cong 0$ (0.1). This is the same order of magnitude as the values of α we have chosen. We have attributed the relatively constant values of α for three compounds RMo_6S_8 (Gd, $\alpha = 0.5$; Tb, $\alpha = 0.4$; and Dy, $\alpha = 0.5$ or 0.6) by analyzing the H_{c2} data. This choice is not unreasonable because α, which is calculated by Eq. (2.13), should be constant for all three compounds except that only the factor $(g_J - 1)[J(J+1)]^{1/2}$ depends explicitly on the rare-earth atoms {Gd, $(g_J - 1)[J(J+1)]^{1/2} = 3.97$; Tb, 3.24; and Dy, 2.66}, because it is not so unnatural to assume that the electronic band structures for the series of these compounds should be nearly the same as a crude approximation. This assumption seems to be consistent with the fact that the antiferromagnetic structures of $TbMo_6S_8$ and $DyMo_6S_8$ are the same,[5] in other words the wave vectors $\vec{Q}$ are the same which should be given by the electronic band structure. Moreover, the constant α ($0.4 \sim 0.6$) means that for all three compounds the area destroyed by the antiferromagnetic energy gap on the Fermi surface is an order of 10% or less compared with the total Fermi surface. This value reminds us of the estimation by Ishikawa,[17] who inferred that "the magnetic electrons," which contribute the RKKY interaction between $4f$ moments, are 30%

of the total conduction electrons by analyzing the magnetic transition temperatures for the series of the heavy rare-earth Chevrel-phase compounds in the Periodic Table.

Throughout this paper we have assumed that the magnetic phase, which is regarded as described by the mean-field theory, is unaffected by the presence of superconductivity, because the formation energy of antiferromagnetism [$\cong O(T_N)$] is much larger than the superconducting condensation energy [$\simeq O(T_c^2/\epsilon_F)$]. As a first approximation we can neglect the effect of superconductivity on the magnetic phase; however, the neutron experiment[5] shows that in $DyMo_6S_8$ the sublattice magnetization of antiferromagnetism does not follow the mean-field-type theory. This is another problem.

Finally, we should point out an interesting possibility, that compounds with relatively large α exhibit superconductivity interrupted by the normal state in the intermediate temperature region. If, in other words, the Fermi surface is anisotropic and the antiferromagnetism can be explained by Fermi-surface nesting condition, then α increases substantially. This condition might be fulfilled in highly anisotropic or "low-dimensional" materials.

ACKNOWLEDGMENTS

We thank Professor M. B. Maple and Dr. M. Ishikawa for their informative correspondence.

APPENDIX A: EVALUATION OF THE INTEGRAL KERNEL $K(p,p',\omega_n)$

A. Diagonal element $K(0,0,\omega_n)$

Substituting Eq. (2.8) into Eq. (2.7), we evaluate $K(0,0,\omega_n)$ of the following form:

$$K(0,0,\omega_n)=2\sum_{k_z>0}\frac{(i\omega_n-\xi_{k-Q})(-i\omega_n-\xi_{k-Q})}{[(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2][(-i\omega_n-\xi_k)(-i\omega_n-\xi_{k-Q})-h_Q^2]}$$
$$+2\sum_{k}\frac{-h_Q^2}{[(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2][(-i\omega_n-\xi_k)(-i\omega_n-\xi_{k-Q})-h_Q^2]}\ , \tag{A1}$$

where we have assumed $Q=(0,0,2k_F)$. If we substruct the term

$$2\sum_{k_z<0}\frac{1}{(i\omega_n-\xi_k)(-i\omega_n-\xi_{-k})}\ , \tag{A2}$$

then we can extend the summation in the first term of Eq. (A1) to all k_z, that is

$$K(0,0,\omega_n)=2\sum_{k}\frac{(i\omega_n-\xi_k)(-i\omega_n-\xi_k)-h_Q^2}{[(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2][(-i\omega_n-\xi_k)(-i\omega_n-\xi_{k-Q})-h_Q^2]}$$
$$-2\sum_{k_z<0}\frac{1}{(i\omega_n-\xi_k)(-i\omega_n-\xi_{-k})}\ . \tag{A3}$$

We define the integral I_K of the first term of Eq. (A3)

$$I_K=\sum_k 2C/AA^*\ , \tag{A4}$$
$$A=t^2-t(2i\omega_n-L^+-L^-)+(i\omega_n-L^+)(i\omega_n-L^-)-h_Q^2\ ,$$
$$C=\tfrac{1}{2}(A+A^*)-t(L^+-L^-)+2\omega_n^2-L^+L^-+(L^-)^2\ ,$$

where we have introduced the cylindrical coordinates (k_ρ, k_ϕ, and k_z), $t=k_\rho^2/2m$, and $L^\pm=k_z^2/2m\pm k_zQ/2m$. After some manipulation we obtain

$$I_K=\frac{2\pi m}{(2\pi)^3}\int_{-\infty}^{\infty}dk_z\int_0^{\infty}dt\left\{\frac{1}{A}+\frac{1}{A^*}+4\left[\omega_n^2+\left(\frac{k_zQ}{2m}\right)^2\right]\frac{1}{AA^*}\right\}$$
$$=\frac{2\pi m}{(2\pi)^3}\int_{-\infty}^{\infty}dk_z\left[\frac{1}{2i\omega_n}\left(1-\frac{h_Q^2}{y^2+\omega_n^2}\right)\ln\frac{\xi_1^*\xi_2^*}{\xi_1\xi_2}-\frac{h_Q^2}{2y(y^2+\omega_n^2)}\ln\frac{\xi_1\xi_1^*}{\xi_2\xi_2^*}\right]\ , \tag{A5}$$

where

$$\xi_{1,2}=i\omega_n-x\pm y,\quad x=k_z^2/2m,\quad y=[(k_zQ/2m)^2+h_Q^2]^{1/2}\ .$$

In view of the fact that ω_n/ϵ_F is very small ($\simeq T_c/\epsilon_F$) the first integral I_1 over k_z in Eq. (A5) is performed as

$$I_1=\frac{2\pi m}{(2\pi)^3}\frac{\pi}{|\omega_n|}(2m)^{1/2}\left[2\sqrt{\beta}-\left(\frac{h_Q^2}{4\epsilon_F}\right)^{1/2}\int_0^{4\epsilon_F\beta/h_Q^2}\frac{dx'}{\sqrt{x'}(x'+1)}+\frac{\omega_n^2}{2\sqrt{\epsilon_F}h_Q}\int_0^{4\epsilon_F\beta/h_Q^2}\frac{dx'}{\sqrt{x'}(x'+1)[x'+(h_Q^2+\omega_n^2)/h_Q^2]}\right] \tag{A6}$$

with $\beta=2\epsilon_F+[(2\epsilon_F)^2+h_Q^2]^{1/2}$. In a similar way the second integral I_2 in Eq. (A5) is given by

$$I_2=\frac{2\pi m}{(2\pi)^3}\left(\frac{m}{2}\right)^{1/2}f\left(\frac{h_Q}{4\epsilon_F},\frac{\omega_n^2}{h_Q^2}\right),$$

$$f\left(\frac{h_Q}{4\epsilon_F},\frac{\omega_n^2}{h_Q^2}\right)=\int_0^\infty\frac{dx'}{(x')^{1/2}(x'+1)^{1/2}[x'+(h_Q^2+\omega_n^2)/h_Q^2]}\ln\left|\frac{(x'+1)^{1/2}+(h_Q/4\epsilon_F)x'}{(x'+1)^{1/2}-(h_Q/4\epsilon_F)x'}\right| \tag{A7}$$

noting that $|h_Q|\ll\epsilon_F$ and retaining the lowest order of $|h_Q|/\epsilon_F$, we finally obtain

$$K(0,0,\omega_n)=\frac{\pi N(0)}{|\omega_n|}\left(1-\frac{\pi|h_Q|}{4\epsilon_F}\right). \tag{A8}$$

B. Off-diagonal element $K(Q,0,\omega_n)$

Then

$$K(Q,0,\omega_n)=\sum_k\frac{(i\omega_n-\xi_{k-Q})h_Q+(-h_Q)(-i\omega_n-\xi_k)}{[(i\omega_n-\xi_k)(i\omega_n-\xi_{k-Q})-h_Q^2][(-i\omega_n-\xi_k)(-i\omega_n-\xi_{k-Q})-h_Q^2]}$$

$$=\sum_k\frac{2h_Q}{AA^*}\left(i\omega_n+\frac{k_zQ}{2m}\right)$$

$$=\frac{2\pi m}{(2\pi)^3}\frac{h_Q}{4}\int_{-\infty}^{\infty}dk_z\left(\frac{1}{y^2+\omega_n^2}\ln\frac{\xi_1^*\xi_2^*}{\xi_1\xi_2}+\frac{i\omega_n}{y(y^2+\omega_n^2)}\ln\frac{\xi_1^*\xi_1}{\xi_2^*\xi_2}\right). \tag{A9}$$

In a similar way, after some manipulation we obtain

$$K(Q,0,\omega_n)=\frac{N(0)}{16\epsilon_F}\left[1+\frac{2i\omega_n}{\pi h_Q}f\left(\frac{h_Q}{4\epsilon_F},\frac{\omega_n^2}{h_Q^2}\right)\right]. \tag{A10}$$

APPENDIX B: A SIMPLE AND INTUITIVE DERIVATION OF THE EFFECTIVE ATTRACTIVE INTERACTION $\tilde{g}N(0)$

Along the direction k_z parallel to $\vec{Q}=(0,0,2k_F)$ the antiferromagnetic energy gap is formed on small area of the Fermi surface (its radius is $2k_F$). This area is estimated as follows: the radius k_ρ of the circle on the Fermi surface, in which the conduction-electron wave functions mix strongly with the wave functions on the Fermi surface of the opposite side to form the antiferromagnetic energy gap, is given by $k_\rho=(2m|h_Q|)^{1/2}$. Then the reduction of the available conduction-electron states due to the formation of the energy gaps is $(4\pi k^2-2\pi k_\rho^2)\,dk=N(0)(1-|h_Q|/2\epsilon_F)d\epsilon$. Therefore, the resultant modification of the effective attractive interaction $\tilde{g}N(0)$ amounts to $\tilde{g}N(0)=gN(0)(1-|h_Q|/2\epsilon_F)$. This intuitive estimation is essentially the correct answer of Eq. (2.12). Now it is easy to understand that the detailed geometry of the Fermi-surface affects the numerical factor of this modification. If we go to low-dimensional systems or highly anisotropic Fermi-surface systems, the weight of this small area to the total Fermi surface becomes substantially increased.

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