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Theory of Superconductors with Overlapping Bands in the Presence of Nonmagnetic Impurities. II

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The transition temperatures of two-band superconductors containing nonmagnetic impurities are further investigated. It is found that because of interband impurity scattering in an impure two-band superconductor, the s-band transition temperature T_{cs} is identical to the d-band transition temperature T_{cd}. That is, for an impure two-band superconductor, there is only one transition temperature. This result is in agreement with the recent experimental findings of Hafstrom and MacVicar. Further, it is noted that the perturbing processes due to the interband impurity scattering are similar to the perturbing processes due to the phonon-exchange interaction between the electrons of the two bands, previously considered by Suhl, Matthias, and Walker.

I. INTRODUCTION

In a previous paper by the present author[1] (hereafter referred to as I), the Green's functions for two-band superconductors containing nonmagnetic impurities have been systematically investigated. Particularly, two limiting cases were separately studied, namely, (a) the intraband phonon coupling limit, $g_s \neq 0$, $g_d \neq 0$, but $g_{sd}=0$, and (b) the interband phonon coupling limit, $g_{sd} \neq 0$, but $g_s = g_d = 0$. As pointed out in Sec. IV of I, for the interband BCS constant g_{sd} to be nonvanishing, the radii of the two Fermi spheres should be identical. Thus, the interband phonon coupling limit is physically less interesting. On the other hand, in the intraband phonon coupling limit, the radii of the two Fermi spheres are not required to be identical. Therefore, this limiting case is relevant to the actual transition metals, such as niobium, in which the d-band Fermi surface is known to be larger than the s-band Fermi surface, although the two Fermi surfaces are not exactly spherical.

In the present paper, we shall only concentrate on the intraband phonon coupling limit, $g_{sd}=0$. As pointed out in I, even in this limiting case, there are unsolved problems. In particular, in I, only the qualitative features of the transition temperatures were studied. In a pure two-band superconductor, in the absence of any kind of interband phonon coupling, there should be two transition temperatures $T_{cs}^{(0)}$ and $T_{cd}^{(0)}$, one corresponding to each of the two bands. Because of larger density of states at the d-band Fermi surface, it is expected that $T_{cd}^{(0)}$ should be larger than $T_{cs}^{(0)}$, and from the point of view of experimentalists, $T_{cd}^{(0)}$ is naturally regarded as the transition temperature of the pure two-band superconductor as a whole, or $T_{cd}^{(0)} \equiv T_c^{(0)}$. In Ref. 2, it was shown that with the existence of interband impurity scattering in an impure two-band superconductor, the d-band transition temperature T_{cd} should be smaller than that of a pure two-band superconductor, $T_{cd} < T_{cd}^{(0)}$. On the other hand, in I, a qualitative conclusion was reached that the s-band transition temperature of an impure two-band superconductor, T_{cs}, should be larger than that of a corresponding pure two-band superconductor, $T_{cs} > T_{cs}^{(0)}$. In Sec. II, we shall show that in an impure two-band superconductor T_{cs} should always be equal to T_{cd}. Once this is shown, we can draw a conclusion that for an impure two-band superconductor, even without interband BCS coupling, there can only be one transition temperature, T_c, which is equal to T_{cd} and T_{cs}.

Although the specific-heat data of niobium superconductors have been successfully explained in terms of the two-band model,[3,4] the recent tunnelling experiments by Hafstrom and MacVicar[5] directly show the formation of distinct s pairs and a pairs in niobium superconductors. The following points should be noted from the Hafstrom-MacVicar experiments. Firstly, the samples measured by Hafstrom and MacVicar should be regarded as impure superconductors. Secondly, in these impure superconductors, no distinct s-band transition temperature T_{cs} is observed, and thus Hafstrom and MacVicar speculated that the s-band transition temperature T_{cs} should coincide with the d-band transition temperature T_{cd}. (See Sec. VI of Ref. 5.) On the basis of the present investigation, we tend to believe that, in an impure two-band superconductor, it is essentially the interband impurity scattering which causes the "interband" coupling. We shall explain this point in detail in Sec. III.

II. TRANSITION TEMPERATURE OF TWO-BAND SUPERCONDUCTORS CONTAINING NONMAGNETIC IMPURITIES IN THE INTRABAND BCS PHONON COUPLING LIMIT

In the intraband phonon coupling limit, $g_{sd}=0$, the Hamiltonian of an impure two-band superconductor is given by

$$\mathcal{H}=\mathcal{H}_0+\mathcal{H}_{\rm int} \quad , \tag{1}$$

$$\begin{aligned}\mathcal{H}_0=&\sum_\sigma \int d^3x\, \psi^\dagger_{s\sigma}(\vec{x})(-\nabla^2/2m_s-\mu)\,\psi_{s\sigma}(\vec{x})\\ &-g_s\int d^3x\, \psi^\dagger_{s\uparrow}(\vec{x})\,\psi^\dagger_{s\downarrow}(\vec{x})\,\psi_{s\downarrow}(\vec{x})\psi_{s\uparrow}(\vec{x})\\ &+\sum_\sigma \int d^3x\, \psi^\dagger_{d\sigma}(\vec{x})(-\nabla^2/2m_d-\mu)\,\psi_{d\sigma}(\vec{x})\\ &-g_d\int d^3x\, \psi^\dagger_{d\uparrow}(\vec{x})\,\psi^\dagger_{d\downarrow}(\vec{x})\,\psi_{d\downarrow}(\vec{x})\,\psi_{d\uparrow}(\vec{x}) \quad ,\end{aligned} \tag{2}$$

$$\begin{aligned}\mathcal{H}_{\rm int}=&\sum_i\sum_\sigma \int d^3x\, V_s(\vec{x}-\vec{R}_i)\,\psi^\dagger_{s\sigma}(\vec{x})\,\psi_{s\sigma}(\vec{x})\\ &+\sum_i\sum_\sigma \int d^3x\, V_d(\vec{x}-\vec{R}_i)\,\psi^\dagger_{d\sigma}(\vec{x})\,\psi_{d\sigma}(\vec{x})\\ &+\sum_i\sum_\sigma \int d^3x\, V_{sd}(\vec{x}-\vec{R}_i)\\ &\times[\psi^\dagger_{s\sigma}(\vec{x})\psi_{d\sigma}(\vec{x})+\psi^\dagger_{d\sigma}(\vec{x})\psi_{s\sigma}(\vec{x})] \quad ,\end{aligned} \tag{3}$$

where $V_s(\vec{x}-\vec{R}_i)$ and $V_d(\vec{x}-\vec{R}_i)$ are the intraband impurity scattering potentials for an impurity located at position $\vec{R}_i$, and $V_{sd}(\vec{x}-\vec{R}_i)$ is the interband impurity scattering potential. The Green's functions for the two bands were obtained in I:

$$G_s(\vec{p}, z_\nu)=\frac{\tilde{z}_{s\nu}+\epsilon_{s\vec{p}}\,\tau_3+\tilde{\Delta}_{s\nu}\,\tau_1}{\tilde{z}^2_{s\nu}-\epsilon^2_{s\vec{p}}-\tilde{\Delta}^2_{s\nu}} \quad , \tag{4}$$

$$G_d(\vec{p}, z_\nu)=\frac{\tilde{z}_{d\nu}+\epsilon_{d\vec{p}}\,\tau_3+\tilde{\Delta}_{d\nu}\,\tau_1}{\tilde{z}^2_{d\nu}-\epsilon^2_{d\vec{p}}-\tilde{\Delta}^2_{d\nu}} \quad , \tag{5}$$

where $\tilde{z}_{s\nu}$ $(\equiv i\tilde{\omega}_{s\nu})$, $\tilde{z}_{d\nu}$ $(\equiv i\tilde{\omega}_{d\nu})$, $\tilde{\Delta}_{s\nu}$, and $\tilde{\Delta}_{d\nu}$ are related to the corresponding quantities z_ν $(\equiv i\omega_\nu)$, Δ_s, and Δ_d of a pure two-band superconductor by the following:

$$\tilde{\omega}_{s\nu}=\omega_\nu+\frac{1}{2\tau_s}\,\frac{\tilde{\omega}_{s\nu}}{(\tilde{\omega}^2_{s\nu}+\tilde{\Delta}^2_{s\nu})^{1/2}}+\frac{1}{2\tau_{sd}}\,\frac{\tilde{\omega}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \,, \tag{6}$$

$$\tilde{\Delta}_{s\nu}=\Delta_s+\frac{1}{2\tau_s}\,\frac{\tilde{\Delta}_{s\nu}}{(\tilde{\omega}^2_{s\nu}+\tilde{\Delta}^2_{s\nu})^{1/2}}+\frac{1}{2\tau_{sd}}\,\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \,, \tag{7}$$

$$\tilde{\omega}_{d\nu}=\omega_\nu+\frac{1}{2\tau_d}\,\frac{\tilde{\omega}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}}+\frac{1}{2\tau_{ds}}\,\frac{\tilde{\omega}_{s\nu}}{(\tilde{\omega}^2_{s\nu}+\tilde{\Delta}^2_{s\nu})^{1/2}} \,, \tag{8}$$

$$\tilde{\Delta}_{d\nu}=\Delta_d+\frac{1}{2\tau_d}\,\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}}+\frac{1}{2\tau_{ds}}\,\frac{\tilde{\Delta}_{s\nu}}{(\tilde{\omega}^2_{s\nu}+\tilde{\Delta}^2_{s\nu})^{1/2}} \,, \tag{9}$$

where $\tau_{s(d)}$ is the intraband relaxation time for the $s(d)$ band, and $\tau_{sd\,(ds)}$ the interband relaxation time for the $s\to d$ $(d\to s)$ processes. Since $N_d(0)\gg N_s(0)$, if all the impurity scattering potentials are assumed to be of the same order in magnitude, we have

$$\frac{1}{2\tau_s}\left(\approx\frac{1}{2\tau_{ds}}\right)\ll\frac{1}{2\tau_d}\left(\approx\frac{1}{2\tau_{sd}}\right) \quad . \tag{10}$$

This inequality shall often be used to simplify various calculations. Its physical meaning is the following. Because of the larger density of states at the d-band Fermi surface, the $s\to d$ impurity scattering processes are more favorable than the $d\to s$ impurity scattering processes; and the d-band electrons tend to be scattered within the d band. Therefore, the interband impurity scattering would influence more the physical properties of the s band than those of the d band.

Based on the above arguments, we can write the first-order approximation for Eqs. (6)–(9) as

$$\tilde{\omega}_{s\nu}\cong\omega_\nu+\frac{1}{2\tau_{sd}}\,\frac{\tilde{\omega}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \quad , \tag{11}$$

$$\tilde{\Delta}_{s\nu}\cong\Delta_s+\frac{1}{2\tau_{sd}}\,\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \quad , \tag{12}$$

$$\tilde{\omega}_{d\nu}\cong\omega_\nu+\frac{1}{2\tau_d}\,\frac{\tilde{\omega}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \quad , \tag{13}$$

$$\tilde{\Delta}_{d\nu}\cong\Delta_d+\frac{1}{2\tau_d}\,\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \quad . \tag{14}$$

Under this approximation, we have

$$\frac{\tilde{\omega}_{d\nu}}{\tilde{\Delta}_{d\nu}}\cong\frac{\omega_\nu}{\Delta_d} \quad . \tag{15}$$

The second-order approximation for $\tilde{\omega}_{d\nu}$ and $\tilde{\Delta}_{d\nu}$ is particularly important, as we shall use it to determine the d-band transition temperature T_{cd}. In the temperature region very close to T_{cd} $(T\gg T^{(0)}_{cs})$, we have $\Delta_s=0$. Therefore, we have the following second-order approximation:

$$\tilde{\omega}_{d\nu}\cong\omega_\nu+\frac{1}{2\tau_d}\,\frac{\tilde{\omega}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}}+\frac{1}{2\tau_{ds}}\,\frac{\omega_\nu}{|\omega_\nu|} \quad , \tag{16}$$

$$\tilde{\Delta}_{d\nu}\cong\Delta_d+\frac{1}{2\tau_d}\,\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}^2_{d\nu}+\tilde{\Delta}^2_{d\nu})^{1/2}} \quad , \tag{17}$$

whence we obtain

$$\frac{\tilde{\omega}_{d\nu}}{\tilde{\Delta}_{d\nu}}\cong\frac{\omega_\nu}{\Delta_d}+\frac{\operatorname{sgn}\nu}{2\tau_{ds}\Delta_d} \quad . \tag{18}$$

[Some subscripts in Eq. (4) of Ref. 2 have been corrected.] When the impurity density is sufficiently low, we obtain $(2\tau_{ds})^{-1}\ll|\omega_\nu|$ for any integer ν [since $\omega_\nu\cong\pi(2\nu+1)T_{cd}$ in this temperature region]. $(2\tau_{ds}\Delta_d)^{-1}$ in Eq. (18) is actually a small quantity as compared with ω_ν/Δ_d. As shown in I, the d-band transition temperature is obtained from the following equation by taking a limit, $T\to T_{cd}$:

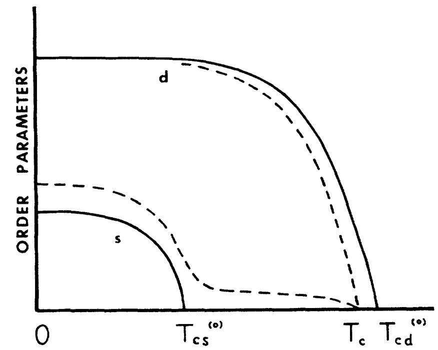

FIG. 1. Effective order parameters for an impure two-band superconductor (dashed curves) plotted against temperature. The order parameters for a corresponding pure two-band superconductor (solid curves) are also plotted for comparison. For an impure two-band superconductor, there can only be one transition temperature, T_c. (The d-band effective order parameter should deviate very little from that oi a pure two-band superconductor.)

$$\ln\frac{T}{T_{cd}^{(0)}}=\frac{2\pi T}{\Delta_d}\sum_{\nu=0}^{\infty}\left(\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}_{d\nu}^2+\tilde{\Delta}_{d\nu}^2)^{1/2}}-\frac{\Delta_d}{\omega_\nu}\right) . \quad (19)$$

[In I, we apply the first-order approximation, Eq. (15), and thus we only reach a crude result $T_{cd} = T_{cd}^{(0)}$.] Now, with the second-order approximation, Eq. (18), we obtain

$$\ln\frac{T_{cd}}{T_{cd}^{(0)}}=\psi(\tfrac{1}{2})-\psi\left(\frac{1}{2}+\frac{1}{4\pi\tau_{ds}T_{cd}}\right) , \quad (20)$$

where $\psi(x)$ is the digamma function. For *low* impurity density, $(2\tau_{ds})^{-1} \ll T_{cd}$, we have

$$T_{cd}\cong T_{cd}^{(0)}-\pi(2\tau_{ds})^{-1} . \quad (21)$$

That is, the d-band transition temperature of a two-band superconductor is lowered by the interband impurity scattering, even though such a lowering is experimentally found to be very small in general.

In I, we have only shown qualitatively that the s-band transition temperature should be raised by the interband impurity scattering. Now, let us look into Eq. (12) in the temperature region $T > T_{cs}^{(0)}$. In this temperature region, Δ_s is identical to zero (note that Δ_s is the s-band order parameter of a two-band superconductor in the absence of impurities), and thus we have

$$\tilde{\Delta}_{s\nu}\cong\frac{1}{2\tau_{sd}}\frac{\tilde{\Delta}_{d\nu}}{(\tilde{\omega}_{d\nu}^2+\tilde{\Delta}_{d\nu}^2)^{1/2}} . \quad (22)$$

Since $\tilde{\Delta}_{d\nu}$ is finite until temperature reaches T_{cd} [from Eq. (22)], we notice that $\tilde{\Delta}_{s\nu}$ would also remain finite until temperature reaches T_{cd}. Thus, we must have T_{cs} and T_{cd} identical. It should be mentioned that with Eq. (I 69) alone, we cannot reach this result; as a matter of fact we can only reach the qualitative result just mentioned.

It should be noticed that in the temperature region $T_{cs}^{(0)} < T \lesssim T_c$ we generally have $\tilde{\Delta}_{s\nu} \ll \tilde{\Delta}_{d\nu}$. It is for this reason that to investigate various physical properties of the impure two-band superconductors, one can nearly let $\tilde{\Delta}_{s\nu} \cong 0$ in this temperature region.[6–8] (In Ref. 7, $T_{c,s}$ should be regarded as $T_{cs}^{(0)}$, according to the notations of the present paper.)

In Fig. 1, the effective s-band and d-band order parameters of an impure two-band superconductor are plotted in comparison with the s-band and d-band order parameters of a corresponding pure two-band superconductor.

III. DISCUSSION

The behavior of the effective order parameters of an impure two-band superconductor shown in Fig. 1 strongly reminds us of the behavior of the order parameters in Fig. 2 of an article by Suhl, Matthias, and Walker (SMW),[9] in which the interband phonon-exchange interaction in a pure two-band superconductor is considered. In the SMW case, instead of our interaction Hamiltonian due to impurity scattering, Eq. (3), the interband phonon-exchange Hamiltonian, is considered. In the $\vec{x}$ space, this SMW Hamiltonian can be written as

$$\mathcal{H}_{exch}=-V_{sd}\int d^3x\,[\psi_{d\uparrow}^{\dagger}(\vec{x})\psi_{d\downarrow}^{\dagger}(\vec{x})\psi_{s\downarrow}(\vec{x})\psi_{s\uparrow}(\vec{x}) + \psi_{s\uparrow}^{\dagger}(\vec{x})\psi_{s\downarrow}^{\dagger}(\vec{x})\psi_{d\downarrow}(\vec{x})\psi_{d\uparrow}(\vec{x})] . \quad (23)$$

In Fig. 2 of SMW, Δ_s and Δ_d are plotted under the condition $V_{sd}^2 \ll g_s g_d$. (Our g_s and g_d are, respectively, V_s and V_d of SMW.) It is noted that when V_{sd} is finite, the two bands also share a common transition temperature T_c (though in the SMW case, $T_c = T_{cd}^{(0)}$). In our Fig. 2, the SMW interband phonon-exchange perturbing process caused by the first term on the right-hand side of Eq. (23) is plotted in the momentum space. The wiggly line denotes the interband phonon-exchange interaction.

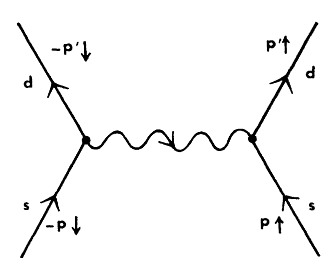

FIG. 2. SMW interband phonon-exchange perturbing process. The wiggly line is to indicate the phonon-exchange interaction.

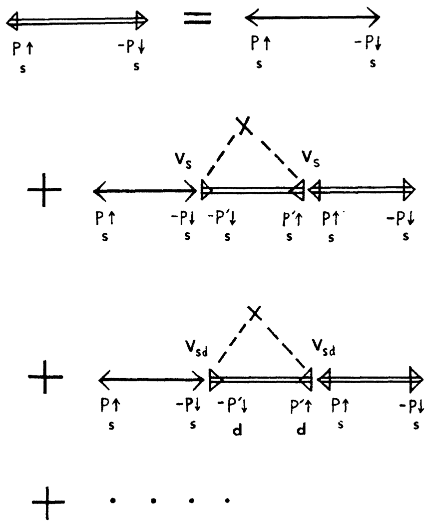

FIG. 3. Dyson equation for the off-diagonal Green's function, $F_{ss}(\vec{p}, Z_\nu)$. The first diagram on the right-hand side is the bare off-diagonal Green's function, $F_{ss}^{(0)}(\vec{p}, Z_\nu)$, the second diagram is due to intraband impurity scattering, and the third diagram is due to interband impurity scattering.

It is interesting that in the case of impurity scattering, one always treats $\mathcal{H}_{\text{int}}$, Eq. (3), in terms of the second-order Born approximation. As a matter of fact, an interband impurity scattering perturbing process, similar to the interband phonon-exchange perturbing process, has already been included in the off-diagonal elements of the 2×2 matrix Green's functions:

$$F_{ss}(\vec{p}, t-t') = -i\langle TC_{s,\vec{p}\uparrow}(t)C_{s,-\vec{p}\downarrow}(t')\rangle \quad , \tag{24}$$

$$F_{dd}(\vec{p}, t-t') = -i\langle TC_{d,\vec{p}\uparrow}(t)C_{d,-\vec{p}\downarrow}(t')\rangle \quad , \tag{25}$$

where $C_{s,\vec{p}\uparrow}$ is the destruction operator for an s electron with momentum $\vec{p}$ and spin up. In Fig. 3, we give the Dyson equation for $F_{ss}(\vec{p}, z_\nu)$ in terms of Feynman diagrams, according to AGD.[10] [As a matter of fact, this is the method we used in I to obtain Eqs. (7) and (9).] On the right-hand side of the equation, only the essential diagrams are explicitly shown. The first diagram is just the bare Green's function, $F_{ss}^{(0)}(\vec{p}, z_\nu)$, the second diagram is due to intraband impurity scattering, and the third diagram is due to interband impurity scattering. It is in the third diagram that we notice the elementary interband impurity scattering perturbing process, which we plot separately in Fig. 4.

Comparing Figs. 2 and 4, we notice the similarity between the SMW interband phonon-exchange perturbing process and the present interband impurity scattering perturbing process. In both processes, a pair of s electrons with opposite spins and momenta are converted to a pair of d electrons with opposite spins and momenta. Actually, owing to this similarity alone, we could already conclude that there can only be one transition temperature associated with an impure two-band superconductor. Indeed, in Ref. 8, we point out that these two perturbing processes compete with each other in two-band superconductors. In a sufficiently dirty two-band superconductor, it is likely that the interband impurity scattering perturbing process would become more important than the SMW interband phonon-exchange perturbing process.

In Fig. 2 of a recent paper by Tang,[11] T_{cs} and T_{cd} for an impure two-band superconductor are treated as being unequal. (It should be pointed out that Tang did not show mathematically that $T_{cs} \neq T_{cd}$.) This is clearly incorrect. Tang's mistake can further be traced back to his Eq. (43). Tang assumes that one can apply the one-band results

$$\omega_\nu \rightarrow \eta_{\omega_\nu}\omega_\nu, \quad \Delta \rightarrow \eta_{\omega_\nu}\Delta \tag{26}$$

(which are valid in converting a pure one-band superconductor to an impure one-band superconductor), to each of the two bands of a two-band superconductor. In view of Eqs. (6)–(9), his assumption is obviously incorrect. It should be emphasized that in I, the present author only draws a qualitative conclusion that "the interband impurity scattering leads to an enhancement of the s-band pair formation and thus leads to an s-band transition temperature which is higher than that of the corresponding pure two-band superconductor" (see p. 472 of I), and does not make a definite statement about the question whether T_{cs} and T_{cd} should be equal. Therefore, Tang's belief that T_{cs} and T_{cd} are not equal has nothing to do with I.

As pointed out earlier in Sec. I, the two-band model, with only intraband phonon coupling considered, has successfully been used to analyze the specific-heat data of impure superconducting transition metals.[3,4] In Ref. 4, it is noted that the s-band order parameter obeys

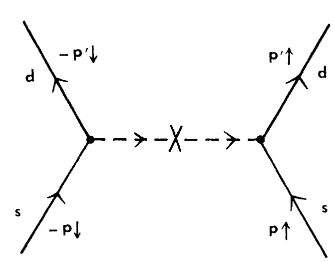

FIG. 4. Interband impurity scattering perturbing process involved in the third diagram in the expansion of Fig. 3. The similarity between Figs. 2 and 4 should be noted.

$$\Delta_s \to \Delta_s + (2\tau_{sd})^{-1} \quad \text{for } T \gtrsim 0, \qquad (27)$$

upon introducing impurities into a two-band superconductor. It is the interband impurity scattering which causes the decrease of the s-band specific heat at low temperatures. The change in Δ_s due to impurity scattering in this temperature region is clearly indicated in Fig. 1. The tunneling experiments by Hafstrom and MacVicar[5] further support the conclusion that there is only one transition temperature associated with an impure two-band superconductor.

Finally, we remark that it is found experimentally that the transition temperature of niobium, T_c, is not sensitive to the amount of impurities present. This can be partly explained by the fact that the lowering of the transition temperature due to the presence of impurity scattering as shown by Eq. (21) is proportional to $(2\tau_{ds})^{-1}$, which is proportional to the small s-band density of states at the Fermi surface, $N_s(0)$. Thus, the illustration of the lowering of the transition temperature T_c from $T_{cd}^{(0)}$ due to the presence of impurities in Fig. 1 should be regarded as qualitative. Further, in the present investigation, we have not taken into account the possible contribution of phonon scattering which might be important for niobium with a transition temperature of the order of 10 °K.

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Heisenberg Ferromagnet with Biquadratic Exchange

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The usual Heisenberg Hamiltonian with bilinear exchange $-2J\,\vec{S}_1\cdot\vec{S}_2$ has been extended to include a biquadratic term $-2\alpha J(\vec{S}_1\cdot\vec{S}_2)^2$, with an adjustable parameter α. A method equivalent to constant coupling was employed to calculate the effect of the biquadratic exchange term on the Curie temperature, magnetization, susceptibility, specific heat, and entropy for lattices with spin-1 atoms. As α goes from 0 to 1, the Curie temperature falls by a factor 2 to 3, while the asymptotic Curie temperature is reduced by the factor 2. The magnetization rises much more rapidly below T_C, and the specific heat has a peak and discontinuity several times higher for $\alpha = 1$. The curvature of the inverse susceptibility increases with α, as does the entropy change taking place above T_C.

I. INTRODUCTION

We will consider the Hamiltonian

$$\mathcal{H} = -2J[\vec{S}_1\cdot\vec{S}_2 + \alpha(\vec{S}_1\cdot\vec{S}_2)^2] - \mu H(S_{1z} + S_{2z}), \quad (1)$$

where J is the Heisenberg exchange integral between neighboring spins S_1 and S_2, with magnetic moments μS_z parallel to an effective (applied plus internal) field H.

For $\alpha = 0$, this is the same as the two-particle Hamiltonian of the form employed by Kasteleijn and van Kranendonk[1] in the constant-coupling approach. For $\alpha = 1$, it is the same as that used by Allan and Betts[2] to investigate the effect of biquadratic exchange on the Curie temperature by means of a high-temperature expansion in powers of reciprocal temperature.

For α small and negative, Joseph[3] also used this Hamiltonian for a high-temperature match of susceptibility data for $KMnF_3$. The need for a small negative biquadratic exchange term was first pointed out by Harris and Owen[4] and Rodbell *et al.*[5] to explain their data on paramagnetic resonance of Mn pairs in MgO. A theoretical basis for the existence of such a term was established by calculations by Anderson[6] and Huang and Orbach[7] of the superexchange interaction in the arrangement Mn-O-Mn.