Possibility of triplet pairing in palladium

D. Fay

*Abteilung für Theoretische Festkörperphysik,
Universität Hamburg, Hamburg, West Germany*

J. Appel

*I. Institut für Theoretische Physik,
Universität Hamburg, Hamburg, West Germany*

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The spin-fluctuation contribution to the p-state pairing interaction in Pd is calculated. The resulting transition temperature, due to spin fluctuations alone, is only of order 10^{-5} K indicating that the occurrence of an observable p-state transition depends strongly on the phonon interaction. Recent calculations of the p-wave phonon interaction are briefly discussed but do not yet allow a reliable prediction of T_c.

The discovery of a condensed phase with triplet pairing in ^{3}He has led to renewed interest in the possibility of finding other p-state superfluids. A particularly attractive candidate is provided by the d electrons in Pd. While the spin-fluctuation (SF) effects in Pd are not as strong as in ^{3}He, they do exist and tend to enhance triplet pairing.

Recently, Foulkes and Gyorffy[1] have calculated, within a spherical Fermi-surface approximation, the p-wave phonon-induced pairing interaction in Pd and find a rather large result: $\lambda_1^{\text{Ph}} \sim 0.2 \lesssim \lambda_0^{\text{Ph}}$, where λ_0^{Ph} is the s-state BCS pairing parameter due to the phonons. The present authors[2] have considered this problem within the site representation[3] employing Doniach's model[4] for the degenerate d subbands. In the site representation, the contact model yields a vanishing contribution to p-state pairing and, therefore, we have calculated the nearest-neighbor interaction as the leading contribution. This corresponds physically to scattering processes where the two electrons are initially separated by a nearest-neighbor distance and where, finally, they again occupy a pair of nearest-neighbor sites. Assuming a short-range phonon Green's function in site space corresponding, roughly speaking, to an Einstein model, we find $|\lambda_1^{\text{Ph}}| << \lambda_0^{\text{Ph}}$, where λ_0^{Ph} is determined by the contact interaction. This should be considered as a preliminary result since the Einstein model is probably not adequate for p-state pairing where the momentum-transfer dependence of the interaction may be important.[5]

In this paper, we discuss in detail only the spin-fluctuation interaction. Our result, namely, that the p-state T_c due to the SF interaction alone is very small ($\lambda_1^{\text{SF}} \lesssim 0.1, T_c \lesssim 10^{-5}$ K), indicates the importance of correctly estimating λ_1^{Ph}. Even a small attractive p-state phonon contribution, when combined with the SF contribution, might yield an observable T_c. Due to the sensitivity of p-state pairing to momentum scattering, however, an extremely clean sample would be necessary.[1]

The contributions of the spin fluctuations to the pairing interaction and to the effective mass are closely related and, as is well known, the original SF theory,[6,7] which employed a one-parameter one-spherical band model for the d holes, overestimates m^*/m in Pd. Inclusion of intersubband (Hund's rule) scattering[4] and, particularly, a momentum-dependent exchange interaction[8] reduce the predicted mass enhancement. We employ essentially the same model-exchange interaction (irreducible particle-hole interaction) as Schrieffer[8] and adjust the model parameters to reproduce the available "experimental" results. The model is then used to calculate λ_1^{SF} and $T_c^{(1)}$. Specifically, we demand that the model yield a Stoner susceptibility enhancement factor[9] $S = 10$, a spin-correlation range[10] $\rho = 5$ Å, and an effective mass[11] $m^*/m = 1 + \delta m/m = 1.7$. Unfortunately, the experimental effective mass contains a phonon contribution. We take $(\delta m/m)^{\text{SF}} = (\delta m/m)^{\text{Ph}}$. This assumption finds some justification on comparison of Pd with Ir where strong SF effects are not present and the phonon interactions should be approximately the same.[11]

The starting point of the T_c calculation is the exact vertex (gap) equation linearized at $T = T_c$,

$$\Gamma_\alpha(\vec{k},\omega_\nu) = -T_c \sum_{\nu'}\sum_{k'}\sum_{\alpha'} I_{\alpha\alpha'}(\vec{k},\omega_\nu;\vec{k}',\omega_{\nu'}) \times G_{\alpha'}(\vec{k}'\omega_{\nu'})G_{\alpha'}(-\vec{k}',-\omega_{\nu'}) \times \Gamma_{\alpha'}(\vec{k}',\omega_{\nu'}) \ . \tag{1}$$

G_α is the exact normal-state single-particle propagator and $I_{\alpha\alpha}(k,k')$ is the irreducible interaction for an antiparallel spin-particle pair scattering from $(k,-k)$ in subband α to $(k',-k')$ in subband α'. Singlet and triplet amplitudes can be obtained as the appropriate combinations of the antiparallel spin amplitude.

We now restrict ourselves to $l=1$ pairing and assume that the p-state contributions of the phonon and short-range Coulomb interactions can be neglected. Then $I_{\alpha\alpha'}$ reduces to $\delta I^{\rm SF}_{\alpha\alpha'}$ which is shown in Fig. 1.[12] The SF contribution to the effective mass is obtained from the self-energy $\Sigma^{\rm SF}_\alpha(k)$ shown in Fig. 2. In Figs. 1 and 2, V is the irreducible particle-hole interaction and the "response function" X consists simply of an iteration of V in the particle-hole channel. It is convenient to define a quantity Λ, which appears in each diagram and is related to the particle-hole t matrix by $t=\Lambda+V$:

$$\Lambda(1,2,3,4) \equiv \sum_{\substack{1',2'\\3',4'}} V(1,2,1',2') \times X(1',2',3',4')V(3',4',3,4) \ . \tag{2}$$

The arguments denote both subband and spin; $1=\alpha_1\sigma_1$, etc., and the convention used is shown in Fig. 3. In the one-parameter one-band paramagnon

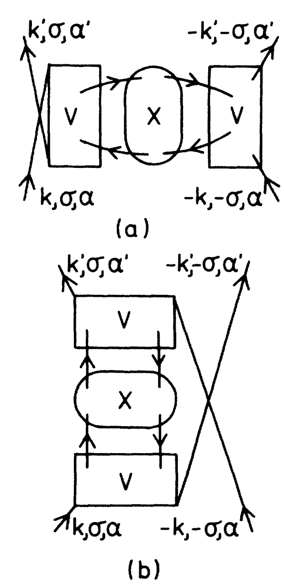

FIG. 1. Spin-fluctuation contributions $\delta I^{\rm SF}_{\alpha\alpha'}(\vec{k},\omega;\vec{k}',\omega')$ to the pair-interaction function for scattering from subband α to subband α'. X represents an iteration of the irreducible interaction V in the particle-hole channel.

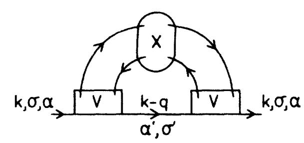

FIG. 2. Self-energy $\Sigma^{\rm SF}_{\alpha,\sigma}(k)$ in subband α.

model the Λ diagrams separate into ladders and strings of bubbles.

It is helpful to first forget the band indices and analyze Λ with respect to spin.[12,13] In Figs. 1 and 2, two combinations of spin indices appear: (a) In $\delta I^{(a)}$ and in Σ, $\Lambda(\sigma,\sigma,\sigma',\sigma')\equiv{}^0\Lambda$ occurs which describes a particle-hole pair with zero z component of spin $(m_s=0)$. ${}^0\Lambda$ includes both singlet and triplet contributions and the diagrams contain both ladders and bubbles. (b) In $\delta I^{(b)}$ and in Σ, $\Lambda(\sigma,-\sigma,\sigma,-\sigma)\equiv{}^1\Lambda$ occurs which is a pure particle-hole triplet $(m_s=\pm1)$ and contains ladder diagrams only. In order to express all contributions in terms of ${}^1\Lambda$, we define

$${}^0\Lambda^\pm \equiv {}^0\Lambda(\sigma,\sigma,\sigma,\sigma) \pm {}^0\Lambda(\sigma,\sigma,-\sigma,-\sigma) \ . \tag{3}$$

One can show[13] that ${}^0\Lambda^-={}^1\Lambda$, i.e., ${}^0\Lambda^-$ is the $m_s=0$ component of the particle-hole triplet. ${}^0\Lambda^+$ is the singlet representing density fluctuations and we will assume that it is included in the short-range screened Coulomb interaction. Neglecting ${}^0\Lambda^+$ with respect to ${}^0\Lambda^-$ and using the above relations we find

$${}^0\Lambda(\sigma,\sigma,\pm\sigma,\pm\sigma) = \pm\tfrac{1}{2}({}^0\Lambda^+ \pm {}^0\Lambda^-) \simeq \tfrac{1}{2}({}^1\Lambda) = \tfrac{1}{2}({}^1V\,{}^1X\,{}^1V) \ . \tag{4}$$

Reinstating the subband indices and using Eq. (4) we obtain

$$\Sigma^{\rm SF}_\alpha(k) = -i\sum_{\alpha'}\int\frac{d^4q}{(2\pi)^4}\frac{3}{2} \times {}^1\Lambda(\alpha,\alpha',\alpha,\alpha';q)G_{\alpha'}(k-q) \ , \tag{5}$$

$$\delta I^{(a)}_{\alpha\alpha'}(k-k') = \tfrac{1}{2}\,{}^1\Lambda(\alpha,\alpha',\alpha',\alpha;k-k') \ , \tag{6}$$

$$\delta I^{(b)}_{\alpha\alpha'}(k+k') = {}^1\Lambda(\alpha,\alpha',\alpha',\alpha;k+k') \ . \tag{7}$$

We now calculate ${}^1\Lambda$ in random-phase-type approximation employing Schrieffer's model[8] which in our notation amounts to setting

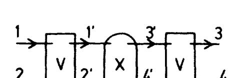

FIG. 3. Diagrammatic convention for the quantity $\Lambda(1,2,3,4)$ which appears in Figs. 1 and 2 and is analyzed in the text.

$$^1V(\alpha_1,\alpha_2,\alpha_3,\alpha_4)=\delta_{\alpha_1\alpha_2}\delta_{\alpha_3\alpha_4}\times{}^1V(\alpha_1,\alpha_2,\alpha_3,\alpha_4;q)\ , \tag{8}$$

where

$$^1V(\alpha,\alpha,\alpha',\alpha';q)=U\delta_{\alpha\alpha'}+J_H(1-\delta_{\alpha\alpha'})+J'_{\alpha\alpha'}(q)\ , \tag{9}$$

$$J'_{\alpha\alpha'}(q)=J'a'^2/(q^2+a'^2)\ . \tag{10}$$

Here, U and J_H are the self-exchange and Hund's-rule exchange *intra*-atomic terms, respectively, and $J'(q)$ is intended to account for the *inter*atomic contributions and the q dependence of the intra-atomic contributions.[8] The form of $J'(q)$ is chosen for simplicity and leads to a range in coordinate space of order $1/a'$. A nice feature of this model is that since $\delta I_{\alpha\alpha'}\propto\delta_{\alpha\alpha'}$, the gap equation is diagonal in the band indices and to obtain δI and Σ only $^1\Lambda(\alpha,\alpha,\alpha,\alpha)$ need be calculated.

For the present purposes a sufficiently accurate estimate of T_c can be obtained by assuming a BCS-type model where $\delta I(\vec{k},\omega;\vec{k}',\omega')=\delta I(\vec{k},\vec{k}')$ for $\omega,\omega'<\omega_{SF}$ and $\delta I=0$ otherwise. Here, $\omega_{SF}\sim E_F/S$ is roughly the maximum paramagnon energy. We assume now Doniach's model[4] for the d-hole subbands. This model consists of six half-spheres at the points $X,-X,Y,-Y,Z,-Z$ in the fcc Brillouin zone or, equivalently, three spheres centered at X, Y, and Z. Taking $|\vec{k}|=|\vec{k}'|\simeq k_F$ and expanding in $\hat{k}\cdot\hat{k}'\equiv\mu$, we obtain

$$T_c^{(1)}\simeq(T_F/S)\exp[-(1+|\lambda_0^{SF}|+\lambda_0^{Ph})/\lambda_1^{SF}]\ , \tag{11}$$

where

$$\lambda_l^{SF}=-\frac{1}{2}\int_{-1}^{+1}P_l(\mu)N_\alpha(0)\delta I_{\alpha\alpha}^{SF}(\mu)\,d\mu \tag{12}$$

in the limit of strong exchange enhancement $\lambda_l<0$ for l even (singlet) and $\lambda_l>0$ for l odd (triplet).[12] The effective mass enhancement due to spin fluctuations is calculated as in Ref. 8 and leads to the result $m^*/m=1+|\lambda_0^{SF}|+\lambda_0^{Ph}$. Taking account of the exchange $\vec{k}'\to-\vec{k}'$ between Eqs. (6) and (7), Eq. (12) becomes

$$\lambda_l^{SF}=-[\tfrac{1}{2}+(-1)^l]\int_0^{2k_F}\frac{q\,dq}{2k_F^2}P_l\left(1-\frac{q^2}{2k_F^2}\right)N_\alpha(0)\times{}^1\Lambda(\alpha,\alpha,\alpha,\alpha;q)\ . \tag{13}$$

Equation (2) for $^1\Lambda(\alpha,\alpha,\alpha,\alpha)$ can easily be diagonalized in the band indices yielding[4,8]

$$^1\Lambda(\alpha,\alpha,\alpha,\alpha;q)=\tfrac{1}{3}[U+2J_H+3J'(q)]^2K_\alpha^+(q)+\tfrac{2}{3}(U-J_H)^2K_\alpha^-(q)\ , \tag{14}$$

with

$$K_\alpha^+(q)=\frac{N_\alpha(0)u(q)}{1-[\bar{U}+2\bar{J}_H+3\bar{J}'(q)]u(q)}\ , \tag{15}$$

$$K_\alpha^-(q)=\frac{N_\alpha(0)u(q)}{1-(\bar{U}-\bar{J}_H)u(q)}\ , \tag{16}$$

where we have defined $\bar{U}=N_\alpha(0)U$, etc. For spherical subbands $u(q)$ is the Lindhard function. We note that in λ_l^{SF} all factors of $N_\alpha(0)$ can be absorbed into the model parameters, thus eliminating the need for an explicit calculation of $N_\alpha(0)$.

The magnetic susceptibility is given by[4,8] $\chi(q)=3K^+(q)$ and the Stoner factor is

$$S=[1-(\bar{U}+2\bar{J}_H+3\bar{J}')]^{-1}\ . \tag{17}$$

The spin-correlation range ρ is obtained by expanding $\chi(q)$ for small q:

$$\chi(q)\underset{q\to0}{\simeq}\frac{3N_\alpha(0)}{1+\rho^2q^2}\ , \tag{18}$$

$$\rho\simeq\left(\frac{S}{12}\right)^{1/2}k_F^{-1}\left(1+\frac{36\bar{J}'k_F^2}{a'^2}\right)^{1/2}\ . \tag{19}$$

We have adjusted the four model parameters $\bar{U}$, $\bar{J}_H$, $\bar{J}'$, and a' in various ways so that we obtain the "experimental" values for Pd; $S\simeq10$, $\rho\simeq5$ Å, and $(\delta m/m)^{SF}\simeq0.35$. Within the spherical subband model, we find in all the parameter combinations investigated that $T_c^{(1)}<3\times10^{-6}$ K.

To go significantly beyond the present model requires enormous numerical effort[10] and leads to results whose validity cannot be easily assessed. In view of this, it seems worthwhile to consider briefly the simplest generalization of the original one-parameter paramagnon theory, namely, replacement of Eq. (9) by

$$^1V(\alpha,\alpha,\alpha',\alpha';q)=U(q)\delta_{\alpha\alpha'}\ , \tag{20}$$

where

$$U(q)=Ua^2/(q^2+a^2)\ . \tag{21}$$

Taking now $k_F=0.9$ Å^{-1} and $E_F\simeq1$ eV as appropriate for the single spherical band model, we find that with $S=10$, a choice of $a=0.707k_F$ fits *both* of the values $|\lambda_0^{SF}|=0.35$ and $\rho=5$ Å. With this choice of a, U has a range in coordinate space of 1.6 Å. The model yields $\lambda_1^{SF}=0.095$ and $T_c^{(1)}=3\times10^{-5}$ K. This two-parameter model must of course be considered as phenomenological but since it does fit correctly the SF mass enhancement and correlation length it may nevertheless provide more reliable results than more sophisticated models.

We note in passing that in the present theory as the ferromagnetic transition is approached ($S\to\infty$), $T_c^{(1)}\to0$ like S^{-1}. Thus $T_c^{(1)}$ should exhibit a maximum as a function of S which we find to occur at about $S = 70$. This situation actually occurs in Ni-Rh alloys[14] and rough calculations yield a p-state T_c on the order of 0.5 K. Unfortunately the p-state superfluid phase in alloys would most likely be destroyed by scattering.[1]

Thus the hope for p-wave pairing in transition metals still seems to rest with Pd, if the phonon interaction is attractive and not vanishingly small.

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