Using a value of 2.7×10^{-3} V for the binding energy of the exciton, an indirect energy gap at 25°C of 0.6643 ± 0.0005 V is obtained.

In addition to these single-phonon transitions, there are also some indications of two-phonon processes at higher energies, as are observed in tunneling experiments,[17] but these are close to the noise level of the experiment. Because of the strong peak associated with the LA phonon transitions, the weak LO phonon is not observed. Its expected location is shown by a dotted arrow. The high-energy sides of the LA associated transition show additional unresolved structure which is probably due to transitions to the higher exciton states.

We are indebted to D. J. Locke for the fabrication of the samples used.

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SUPERCONDUCTIVITY IN FERROMAGNETIC ALLOYS

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(Received 31 August 1965)

In a ferromagnetic alloy, the spins of the magnetic impurities are correlated and not free to rotate. Then the exchange interaction between conduction electrons and magnetic impurities involves energy transfer. Therefore the theory proposed by Gor'kov and Rusinov[1] on superconductivity in ferromagnetic alloys assuming a static exchange interaction is extended by using a time-dependent exchange interaction between conduction electrons and magnetic impurities.[2] The system of impurity spins is treated by quantum field-theoretical methods using the special technique developed by Abrikosov.[3] The electron-electron interaction resulting from the exchange of magnons is included in the effective electron-electron interaction. The electronic self-energy resulting from the dynamic electron–spin-wave coupling is determined analogously as in the case of dynamic electron-phonon coupling by Éliashberg.[4] The dimensionless electron–magnetic-impurity exchange coupling constant turns out to be much bigger than the dimensionless electron-phonon coupling constant.

There is no indication so far that the anomalous electronic scattering derived by Kondo[5] for exchange coupling between conduction electrons and magnetic impurities is significant for superconducting paramagnetic alloys. This anomalous electronic scattering decreases with decreasing degeneracy of the impurity-spin energy levels and increasing Zeeman energy of the impurity spins and thus should become even less important for superconducting ferromagnetic alloys. Therefore it should be possible in any case to isolate effects arising from Kondo's anomalous electronic scattering from the effects arising from electron–spin-wave coupling and thus for simplicity the anomalous electronic scattering is neglected. Also for simplicity, the electron-phonon coupling is treated using the BCS approximation.

We show that the ferromagnetic ordering of the magnetic impurities in a superconductor is strongly reflected in the density of states of the conduction electrons and thus could be detected by tunneling experiments.

The exchange coupling between conduction electrons and magnetic impurities is treated using the special field-theoretical methods developed by Abrikosov[3] and using Migdal's approximation for the electron-spin-wave vertex function. In order to include the magnetic field $\vec{H}$ and the associated vector potential $\vec{A}$ resulting from the magnetic impurities, it is assumed that the mean free path of an electron is much smaller than the inverse of the order parameter of the superconductor and also that the order parameter is space independent.[6] Spin-orbit scattering of the electrons due to nonmagnetic impurities, crystal inhomogenities, and boundaries is taken into account. One obtains then from Gor'kov's equations, after averaging over the lattice positions of the impurities, the spin-dependent thermal electronic Green's functions

$$G_{\pm}(\vec{p},\omega_n)=-\frac{i\tilde{\omega}_{\pm}+\xi_p+(e/m)\vec{p}\cdot\vec{A}}{\Omega_{\pm}(\vec{p},\omega_n)},\tag{1}$$

and

$$F_{\pm}^{+}(\vec{p},\omega_n)=-i\frac{\tilde{\Delta}_{\pm}}{\Omega_{\pm}(p,\omega_n)},\tag{2}$$

where

$$\Omega_{\pm}(\vec{p},\omega_n)=[\tilde{\omega}_{\pm}-i(e/m)\vec{p}\cdot\vec{A}]^2+\tilde{\Delta}_{\pm}^{\,2}+\xi_p^{\,2},\quad \omega_n=(2n+1)\pi T,\tag{3}$$

$$\tilde{\omega}_{\pm}=\omega_n\mp i\mu_B(H+I)+\frac{1}{2\tau}\int\frac{d\Omega}{4\pi}\frac{\tilde{\omega}_{\pm}-i(e/m)\vec{p}\cdot\vec{A}}{\{[\tilde{\omega}_{\pm}-i(e/m)\vec{p}\cdot\vec{A}]^2+\tilde{\Delta}_{\pm}^{\,2}\}^{1/2}}$$
$$+\frac{1}{3\tau_2}\int\frac{d\Omega}{4\pi}\frac{\tilde{\omega}_{\mp}-i(e/m)\vec{p}\cdot\vec{A}}{\{[\tilde{\omega}_{\mp}-i(e/m)\vec{p}\cdot\vec{A}]^2+\tilde{\Delta}_{\mp}^{\,2}\}^{1/2}}+\Sigma_{\mp}^{(1)}(\omega_n),\tag{4}$$

$$\tilde{\Delta}_{\pm}=\Delta+\left(\frac{1}{2\tau}-\frac{1}{\tau_1}\right)\int\frac{d\Omega}{4\pi}\frac{\tilde{\Delta}_{\pm}}{\{[\tilde{\omega}_{\pm}-i(e/m)\vec{p}\cdot\vec{A}]^2+\tilde{\Delta}_{\pm}^{\,2}\}^{1/2}}$$
$$+\frac{1}{3\tau_2}\int\frac{d\Omega}{4\pi}\frac{\tilde{\Delta}_{\mp}}{\{[\tilde{\omega}_{\mp}-i(e/m)\vec{p}\cdot\vec{A}]^2+\tilde{\Delta}_{\mp}^{\,2}\}^{1/2}}-\Sigma_{\mp}^{(2)}(\omega_n),\tag{5}$$

and

$$\frac{1}{\tau}=\frac{1}{\tau_0}+\frac{1}{\tau_1}+\frac{1}{\tau_2},\quad I=n_1S\langle\mathcal{G}\rangle.\tag{6}$$

The normal-state electronic energy ξ_p is measured with respect to the Fermi energy. The electronic transport collision times τ_0, τ_1, and τ_2 result from electron scattering due to all impurities (neglecting exchange and spin-orbit coupling), from static exchange scattering due to the impurity-spin component S_z in the direction of $\vec{I}$, and from spin-orbit scattering, respectively.[1] $\vec{I}$ denotes the average exchange field which results from the ferromagnetically ordered system of magnetic impurities of density n_1 with spin S per impurity and average exchange-coupling matrix element $\langle\mathcal{G}\rangle$. Note that in the paramagnetic state, $\vec{H}=\vec{I}=0$ in the absence of molecular fields. In evaluating $\vec{p}\cdot\vec{A}$, $\vec{p}$ is taken as lying on the Fermi surface and $|\vec{A}|^2$ is approximated by $(1/\Omega)\int d^3r\,|\vec{A}(\vec{r})|^2$.[6] The spin-dependent self-energies

$$\Sigma_{\pm}^{(1)}(\omega_n)=iT\sum_{n'=-\infty}^{\infty}\int\frac{d^3p'}{(2\pi)^3}G_{\pm}(\vec{p}\,',\omega_n)\times E(\vec{p}-\vec{p}\,',i\omega_n-i\omega_{n'}),\tag{7}$$

and

$$\Sigma_{\pm}^{(2)}(\omega_n)=iT\sum_{n'=-\infty}^{\infty}\int\frac{d^3p'}{(2\pi)^3}F_{\pm}^{+}(\vec{p}',\omega_n)\times E(\vec{p}-\vec{p}',i\omega_n-i\omega_{n'}), \tag{8}$$

result from the dynamic electron–spin-wave coupling applying Migdal's approximation to the vertex function. $E(\vec{q},i\omega_n)$ denotes the Fourier-transformed thermal impurity-spin propagator describing the fluctuation of the impurity spins. $\Sigma_{\pm}^{(1)}$ and $\Sigma_{\pm}^{(2)}$ arise from virtual electron-scattering processes involving emission and absorption of the same spin wave. Using the spectral representation for the spin-correlation function E and assuming that the resulting spectral density function $L_\lambda(\vec{q},\omega_n)$ for impurity-spin excitations of kind λ is antisymmetric in ω_n, one obtains

$$\Sigma_{\pm}^{(1)}(\omega_n)=\int_{-\infty}^{\infty}d\omega'\int\frac{d\Omega}{4\pi}K(\omega_n,\omega')\,\mathrm{sign}\omega'\,\mathrm{Re}\left\{\frac{\tilde{\omega}_{\pm}(\omega')+(e/m)\vec{p}\cdot\vec{A}}{\{[\tilde{\omega}_{\pm}(\omega')+(e/m)\vec{p}\cdot\vec{A}]^2-\tilde{\Delta}_{\pm}^{2}(\omega')\}^{1/2}}\right\}, \tag{9}$$

and

$$\Sigma_{\pm}^{(2)}(\omega_n)=\int_{-\infty}^{\infty}d\omega'\int\frac{d\Omega}{4\pi}K(\omega_n,\omega')\,\mathrm{sign}\omega'\,\mathrm{Re}\left\{\frac{\tilde{\Delta}_{\pm}(\omega')}{\{[\tilde{\omega}_{\pm}(\omega')+(e/m)\vec{p}\cdot\vec{A}]^2-\tilde{\Delta}_{\pm}^{2}(\omega')\}^{1/2}}\right\}, \tag{10}$$

where

$$K(\omega_n,\omega')\equiv\frac{N(0)}{4p_0^{\,2}}\int_0^\infty dz\left(\frac{\tanh(\omega'/2T)-\cosh(z/2T)}{\omega'-i\omega_n-z-i\delta}-\frac{\tanh(\omega'/2T)+\cosh(z/2T)}{\omega'-i\omega_n+z-i\delta}\right)\times\int_0^{q_c}dq\,q\sum_\lambda L_\lambda(q,z)\,|g_{q,\lambda}(z)|^2, \tag{11}$$

$$|g_{q,\lambda}(\omega)|^2=2n_1S\,|\mathcal{G}_\lambda(q,\omega)|^2, \tag{12}$$

and

$$q_c=\min(2p_0,q_{\max}). \tag{13}$$

$N(0)$ is the density of states at the Fermi surface. $\mathcal{G}_\lambda(q,\omega)$ denotes the exchange-coupling matrix element taking into account screening of the exchange interaction between conduction electrons and magnetic impurities. Note that the expressions for $\Sigma_{\pm}^{(1)}$ and $\Sigma_{\pm}^{(2)}$ given by Eqs. (9) and (10), respectively, reduce in the limit of static exchange coupling to the expressions given by Gor'kov and Rusinov[1] for $\Sigma_{\pm}^{(1)}$ and $\Sigma_{\pm}^{(2)}$. Also note that the energy shift and the anomalous density of states for electrons in dilute magnetic alloys found by Kondo[2] can be rederived using Eqs. (9) and (11).

It follows from Eqs. (4) and (5) that

$$\frac{\omega_n\mp i(\mu_BH+I)}{\Delta}=u_{\pm}\left(1-\frac{\beta}{(1+u_{\pm}^{2})^{1/2}}\right)-\frac{1}{3\tau_2\Delta}\frac{u_{\mp}-u_{\pm}}{(1+u_{\mp}^{2})^{1/2}}-\frac{1}{\Delta}[u_{\pm}\Sigma_{\mp}^{(2)}(\omega_n)+\Sigma_{\mp}^{(1)}(\omega_n)], \tag{14}$$

where

$$u_{\pm}\equiv\frac{\tilde{\omega}_{\pm}}{\tilde{\Delta}_{\pm}},\quad \beta\equiv\frac{1}{\tau_1\Delta}+\frac{2\alpha}{\Delta}\left(\frac{1}{\tau}-\frac{2}{\tau_1}\right)^{-1},\quad \alpha\equiv\left\langle\left(\frac{e}{m}\vec{p}\cdot\vec{A}\right)^2\right\rangle=\frac{1}{3}\left(\frac{ep_0}{m}\right)^2\frac{1}{\Omega}\int d^3r\,|\vec{A}(\vec{r})|^2. \tag{15}$$

For $S\gg1$ and electronic energies ω bigger than $\omega_0+2\omega_1$, where ω_0 is the gap in the electronic energy excitation spectrum and ω_1 is the average energy of a magnon, β is approximately given by

$$\beta = \frac{\Delta_{00}}{2\Delta}\left[\frac{n_1}{n_c} + \left(\frac{H}{H_{c0}}\right)^2\right].$$

Δ_{00} is the energy gap at zero temperature of the pure superconductor, n_c denotes the concentration of magnetic impurities which destroys superconductivity, and H_{c0} is the critical magnetic field at zero temperature. Note that in general $u_\pm(\omega_n)$ must be determined numerically. However, for energies ω either of the order of the energy gap or such that $\omega/\Delta \gg 1$, one can find an approximate analytical solution for $u_\pm(\omega_n)$.

Since a knowledge of $u_\pm(\omega_n)$ suffices to determine the thermodynamic and electrodynamic properties of any superconductor, Eq. (14), with Eqs. (9) and (10), constitutes the basic mathematical formulation of the theory of superconducting ferromagnetic alloys. In particular, the ratios N_s^+/N_n^+ and N_s^-/N_n^- [of the superconducting-state density of states to the normal-state density of states for electrons whose spin is parallel to $\vec{\mathrm{I}}$ and antiparallel to $\vec{\mathrm{I}}$, respectively] are given by the equation

$$\frac{N_s^\pm(\omega)}{N_n^\pm(0)} = \mathrm{Im}\left\{\frac{u_\pm(\omega)}{[1-u_\pm^2(\omega)]^{1/2}}\right\}. \tag{16}$$

$u_\pm(\omega)$ is given by Eq. (14), replacing $i\omega_n$ by ω and $u_\pm(\omega_n)$ by $-iu_\pm(\omega)$.

The density of states is now calculated for zero temperature, neglecting small and, for the structure in the density of states, unessential quantities of order $(\mu_B H + I)/\omega$ in Eq. (14) so that $u_+ = u_- = u$, and approximating the spectral density function $L_2(q_1\omega)$ of the spin correlation function representing the impurity-spin fluctuation by one Lorentzian function of width $\omega_2 = 0.2\omega_1$ centered at the average spin-wave excitation energy $\omega_1 = T_K/S$, where T_K denotes the Curie temperature. It follows then from Eqs. (9) and (10)

$$\Sigma_\pm^{(1)}(\omega) = \lambda \int_{\omega_0}^\infty d\omega' \,\mathrm{Im}\left\{\frac{u(\omega')}{[1-u^2(\omega')]^{1/2}}\right\} K_+(\omega,\omega'), \tag{17}$$

$$\Sigma_\pm^{(2)}(\omega) = \lambda \int_{\omega_0}^\infty d\omega' \,\mathrm{Im}\left\{\frac{1}{[1-u^2(\omega')]^{1/2}}\right\} K_-(\omega,\omega'), \tag{18}$$

where

$$K_\pm(\omega,\omega') \equiv \frac{\omega_1 - i\omega_2}{2}\left(\frac{1}{\omega'-\omega+\omega_1-i\omega_2} \mp \frac{1}{\omega'+\omega+\omega_1-i\omega_2}\right), \tag{19}$$

and

$$\lambda = \frac{2n_1 SN(0)\,|\mathcal{G}|^2}{\omega_1} = \frac{3}{\gamma}, \quad \omega_1 = \tfrac{2}{3}\gamma n_1 SN(0)\,|\mathcal{G}|^2. \tag{20}$$

λ is the dimensionless electron–spin-wave coupling constant. Note that the screening of the electron–spin-wave interaction is taken into account exactly. The constant γ is of the order of unity.[2] Figure 1 shows the structure in the density of states resulting from the dynamic exchange interaction between conduction electrons and magnetic impurities. The curve parameters are consistent with the phase diagram obtained by Suhl, Matthias, and Corenzwit.[7]

The structure obtained should be detectable by tunneling experiments. Note that for a given concentration of magnetic impurities the change in the density of states due to the dynamic electron–spin-wave interaction increases with increasing Curie temperature. Further note that the structure in the density of states arising from the dynamic electron-phonon interaction is important only at energies much higher than the energies for which the structure in the density of states due to electron–spin-wave coupling is important.

The structure in the electronic density of states is characteristic for the spin-wave field

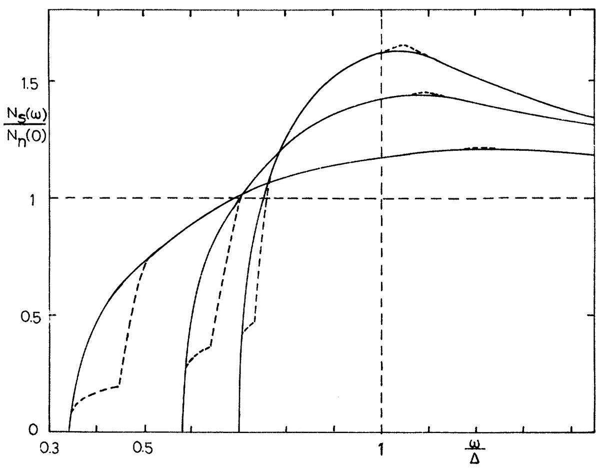

FIG. 1. The solid curves show the electronic density of states of superconducting alloys using a static electron-impurity exchange interaction. The dashed lines show the change in the electronic density of states as resulting from a dynamic electron–spin-wave interaction in magnetic alloys. The spectrum of spin-wave excitations is given by one Lorentzian function. From left to right the curves shown correspond to alloys for which $n_1/n_c = 0.55$, $T_K = 3°K$, $w_1/\Delta = 0.13$; $n_1/n_c = 0.30$, $T_K = 1.8°K$, $w_1/\Delta = 0.07$; and $n_1/n_c = 0.19$, $T_K = 0.90°K$, $w_1/\Delta = 0.03$, respectively. n_1 and n_c denote the concentration of magnetic impurities and the concentration of magnetic impurities which destroy superconductivity, respectively. w_1 denotes the average energy of a spin-wave excitation. The area removed from the solid curves for $w/\Delta < 1$ is equal to that added for $w/\Delta > 1$, but the latter is more spread out. At sufficiently low temperatures the second derivative of the tunneling current with respect to the voltage should exhibit peaks at w_0 and at approximately $w_0 + w_1$.

and thus can be used to obtain information about the spectrum of the impurity–spin-wave excitations. A complicated, multipeaked spectrum of the impurity–spin-wave excitations might be approximated theoretically by a sum of Lorentzian functions.

Using an appropriate form for the fields $\vec{H}$ and $\vec{I}$ and for the spectral density function representing the impurity-spin fluctuation, the theory outlined above is also applicable in the case of antiferromagnetic alloys and if the impurity spins are fixed at low temperatures by an internal field due to crystalline anisotropy or by a molecular field whose direction and magnitude may vary for each magnetic impurity.

In the tunneling experiments by Reif and Woolf,[8] the paramagnetic impurity spins might feel only a small molecular field and, consequently, the resulting structure in the density of states will be small.

It is a pleasure to thank Professor J. W. Garland, Jr., for many helpful suggestions and discussions, and Professor Sir Nevill F. Mott and Dr. V. Heine for their hospitality.

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