THEORY OF s-f EXCHANGE FOR NONVANISHING ORBITAL ANGULAR MOMENTA

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s-f-exchange interaction is considered in the case when the total angular f-shell momentum J is a good quantum number. The Hamiltonian for s-f-exchange connected with the changes in J is established and its effect on indirect exchange interaction and superconductivity is analyzed. The results obtained by Brout and Suhl[7] and by Mattis[9] are critically discussed.

1. In an analysis of s-f exchange in rare-earth metals it is necessary to take into account in the general case (with the exception of gadolinium) not only the spin **S** of the unfilled s-shell, but also its non-zero orbital angular momentum **L**. De Gennes[1] advanced the hypothesis that in these metals the s-f exchange is determined not by the operator **S**, but by the operator $(g-1)\mathbf{J}$, where g is the Landé factor. This hypothesis signifies intuitively that only the **S** component parallel to the total angular momentum of the shell $\mathbf{J} = \mathbf{L} + \mathbf{S}$ is effective in the exchange[2]. The de Gennes hypothesis was later corroborated by Liu[3], who obtained, confining himself to the principal term of the expansion of the phase factor $\exp{(i\mathbf{k}\cdot\mathbf{r})}$ of the conduction-electron wave functions in spherical harmonics, the following expression for the s-f exchange Hamiltonian:

$$H = -I(\mathbf{k}, \mathbf{k}')[\tfrac{1}{2}N + (g-1)\mathbf{sJ}], \qquad (1)$$

where N is the number of electrons in the f-shell, and **s** is the spin of the conduction electron.

The indirect exchange due to a Hamiltonian of the type (1) was considered by many authors[4,5]. Kaplan and Lyons[5] have shown that inclusion of the expansion term following the principal one leads to the appearance of anisotropic exchange interaction, besides the Heisenberg-type exchange $\sim \mathbf{J}_n\mathbf{J}_m$.

A Hamiltonian of the type (1) describes s-f exchange in which the quantum number J is conserved. There exist, however, phenomena whose analysis calls for inclusion of the s-f exchange accompanied by variations of J. For example, the lowering of the critical temperature T_c of superconducting lanthanum following addition of Eu impurity[6] cannot be explained if a Hamiltonian of type (1) is used, since J = 0 for Eu in the ground state. In this connection, Brout and Suhl[7] expressed the opinion

that the influence of the Eu impurity on T_c of lanthanum is due to s-f exchange in which J of the Eu f-shell changes by unity. However, a quantitative estimate of the lowering of T_c, based on Herring's assumption[8] that the free energies of the normal and superconducting states come closer together in the presence of magnetic impurities, has led to an anomalously small value for the energy Δ connected with unity of change J. Namely Brout and Suhl[7] obtained for Eu a value $\Delta \sim 20-40$ cm^{-1}, whereas usually $\Delta \sim 10^3$ cm^{-1} for rare earths. They have also indicated[7] that the s-f exchange accompanied by changes in J may possibly influence the establishment of ferromagnetic order.

Kaplan and Lyons[5] believe, to the contrary, that the transitions accompanied by changes in J cannot make any contribution to the isotropic exchange for different atoms. Recently, however, Mattis[9] concluded from the results of[7] that exchange connected with changes in J can lead to an indirect exchange of the type $\sim \mathbf{J}_m\mathbf{J}_n$ which has, along with an oscillating term of the Ruderman-Kittel type, also a non-oscillating term. Thus, unlike the case when the quantum number J is assumed to be conserved, in the theory of s-f exchange accompanied by changes in J there are several mutually contradictory statements. In this connection, it is of interest to consider the s-f exchange which we developed by an earlier method[10], connected with changes in J, and also to assess its influence on indirect exchange and on superconductivity; this is the purpose of the present article.

2. The s-f exchange interaction Hamiltonian has in the second-quantization representation the following[10]:

$$H = -\sum_{\mathbf{k}, \mathbf{k}', n, \sigma_1, \sigma_2, \mu} I_{\mu, \mathbf{k}, \mathbf{k}', n} a^*_{n\mu\sigma_1} a_{n\mu\sigma_2} a^*_{\mathbf{k}'\sigma_2} a_{\mathbf{k}\sigma_1}, \qquad (2)$$

where $a^*_{n\mu\sigma}$ and $a^*_{k\sigma}$ (and also $a_{n\mu\sigma}$ and $a_{k\sigma}$) are the Fermi electron creation (annihilation) operators, n is the number of the ion of the rare-earth element, μ is the magnetic quantum number in the f-state, σ is the electron spin projection, and the exchange integral $I_{\mu,k,k',n}$ has the following form:

$$I_{\mu,k,k',n} = \exp\{i(k-k')R_n\}I_\mu(k,k'). \quad (3)$$

Here $I_\mu(k,k')$ does not depend on the position R_n of the rare-earth ion, and is determined by

$$I_\mu(k,k') = e^2 \int \frac{\varphi_{k'}^*(r_1)\Phi_{\lambda\mu}^*(r_1)\varphi_k(r_2)\Phi_{\lambda\mu}(r_2)}{|r_1-r_2|} dr_1\,dr_2, \quad (3')$$

where φ_k is the wave function of the conduction electron, and $\Phi_{\lambda\mu}$ is the atomic wave function of the f-state with $\lambda = 3$.

Introducing the spin operators

$$S_{n\mu}^z = N_{n\mu\uparrow} - {}^1/_2 N_{n\mu} = {}^1/_2 N_{n\mu} - N_{n\mu\downarrow},$$

$$S_{n\mu}^+ = a^*_{n\mu\uparrow} a_{n\mu\downarrow}, \quad S_{n\mu}^- = a^*_{n\mu\downarrow} a_{n\mu\uparrow}, \quad (4)$$

where $N_{n\mu\sigma}$ is the number of electrons in the state $\Phi_{n\lambda\mu\sigma}$, and $N_{n\mu} = N_{n\mu\uparrow} + N_{n\mu\downarrow}$, and summing over σ_1 and σ_2, we transform (2) into

$$H = -\sum_{k,k',n,\mu} I_{\mu,k,k',n}\{{}^1/_2 N_{n\mu}(a^*_{k'\uparrow}a_{k\uparrow} + a^*_{k'\downarrow}a_{k\downarrow}$$
$$+ (a^*_{k'\uparrow}a_{k\uparrow} - a^*_{k'\downarrow}a_{k\downarrow})S_{n\mu}^z + a^*_{k'\downarrow}a_{k\uparrow}S_{n\mu}^+ + a^*_{k'\uparrow}a_{k\downarrow}S_{n\mu}^-\}. \quad (5)$$

Using identity (10) from [10], we can write (5) in the form of two sums:

$$H = -\sum_{k,k',n} \bar{I}_{k,k',n}\{{}^1/_2 N_n(a^*_{k'\uparrow}a_{k\uparrow} + a^*_{k'\downarrow}a_{k\downarrow})$$
$$+ (a^*_{k'\uparrow}a_{k\uparrow} - a^*_{k'\downarrow}a_{k\downarrow})S_n^z$$
$$+ a^*_{k'\downarrow}a_{k\uparrow}S_n^+ + a^*_{k'\uparrow}a_{k\downarrow}S_n^-\}$$
$$- \frac{1}{2\lambda+1}\sum_{k,k',n}\sum_{\mu_1<\mu_2}(I_{\mu_1,k,k',n} - I_{\mu_2,k,k',n})$$
$$\times \{{}^1/_2(N_{n\mu_1} - N_{n\mu_2})(a^*_{k'\uparrow}a_{k\uparrow} + a^*_{k'\downarrow}a_{k\downarrow})$$
$$+ (a^*_{k'\uparrow}a_{k\uparrow} - a^*_{k'\downarrow}a_{k\downarrow})(S_{n\mu_1}^z - S_{n\mu_2}^z)$$
$$+ a^*_{k'\downarrow}a_{k\uparrow}(S_{n\mu_1}^+ - S_{n\mu_2}^+) + a^*_{k'\uparrow}a_{k\downarrow}(S_{n\mu_1}^- - S_{n\mu_2}^-)\}, \quad (6)$$

where the first sum depends on the average exchange integral

$$\bar{I}_{k,k',n} = \frac{1}{2\lambda+1}\sum_\mu I_{\mu,k,k',n},$$

on the total spin $S_n = \sum_\mu S_{n\mu}$, and on the number N_n of f-shell electrons, while the second sum contains combinations of exchange integrals and spin operators which are anti-symmetrical with respect to μ_1 and μ_2.

To simplify the subsequent analysis, we confine

ourselves here to an account of only the "principal" terms (see [3,5]) and we assume that $I_{\mu,k,k',n}$ does not depend on μ. Then the second sum in (6) vanishes, and in the first sum we use the identity

$$S \equiv \alpha_J J + Q, \quad (7)$$

where $a_J = g_J - 1$ and by definition $Q = S - \alpha_J J$. We then get in place of (6)

$$H = H_1 + \tilde{H}_1; \quad (8)$$

$$H_1 = -\sum_{k,k',n} I_{k,k',n}\{{}^1/_2 N_n(a^*_{k'\uparrow}a_{k\uparrow} + a^*_{k'\downarrow}a_{k\downarrow})$$
$$+ (a^*_{k'\uparrow}a_{k\uparrow} - a^*_{k'\downarrow}a_{k\downarrow})\alpha_J J_n^z$$
$$+ a^*_{k'\downarrow}a_{k\uparrow}\alpha_J J_n^+ + a^*_{k'\uparrow}a_{k\downarrow}\alpha_J J_n^-\}, \quad (9)$$

$$\tilde{H}_1 = -\sum_{k,k',n} I_{k,k',n}\{(a^*_{k'\uparrow}a_{k\uparrow} - a^*_{k'\downarrow}a_{k\downarrow})Q_n^z$$
$$+ a^*_{k'\downarrow}a_{k\uparrow}Q_n^+ + a^*_{k'\uparrow}a_{k\downarrow}Q_n^-\}. \quad (10)$$

It is easy to see that the Hamiltonian H_1 in (9) is equivalent to the Hamiltonian (1). Therefore the expression obtained by us for H_1 gives, in our opinion, the simplest and most direct justification of the de Gennes hypothesis [1]. However, since we are interested in effects not accounted for by the Hamiltonian (1) or (9), we turn to the analysis of the Hamiltonian $\tilde{H}_1$, which was not considered earlier. To clarify the role of $\tilde{H}_1$, it is necessary to establish the meaning of the operator Q. Taking into account the definition of α_J, it is easy to verify that the operator Q is orthogonal to the operator J. Therefore the diagonal matrix elements of Q in states with definite J are equal to zero. Furthermore, since the following commutation relations hold:

$$[J^i Q^k]_- = ie_{ikl}Q^l, \quad (11)$$

where e_{ikl} is an antisymmetrical unit tensor, we obtain, by using formulas (27.11) and (27.13) from [2], the following nonvanishing matrix elements of Q:

$$\langle J-1, J^z|Q^z|J, J^z\rangle = A_{J-1}^J[J^2 - (J^z)^2]^{1/2},$$

$$\langle J+1, J^z|Q^z|J, J^z\rangle = A_{J+1}^J[(J+1)^2 - (J^z)^2]^{1/2},$$

$$\langle J-1, J^z+1|Q^+|J, J^z\rangle = A_{J-1}^J[(J-J^z)(J-J^z-1)]^{1/2},$$

$$\langle J+1, J^z+1|Q^+|J, J^z\rangle = -A_{J+1}^J[(J+J^z+1)(J+J^z+2)]^{1/2},$$

$$\langle J-1, J^z-1|Q^-|J, J^z\rangle = -A_{J-1}^J[(J+J^z-1)(J+J^z)]^{1/2},$$

$$\langle J+1, J^z-1|Q^-|J, J^z\rangle = A_{J+1}^J[(J-J^z+1)(J-J^z+2)]^{1/2}. \quad (12)$$

We see from (12) that Q^z, Q^+, and Q^- change J^z by 0, +1, and -1, respectively. However, these operators change J by only ±1. It follows therefore that

the Hamiltonian $\tilde{H}_1$ describes the s-f exchange accompanied by a change $\pm\hbar$ in the total angular momentum of the f-shell.

The coefficients $A^J_{J\pm1}$ in (12) can be determined from the system of equations

$$(A^J_{J-1})^2(2J-1)J + (A^J_{J+1})^2(2J+3)(J+1)$$
$$= S(S+1) - a_J^2 J(J+1),$$

$$(A^J_{J-1})^2(2J-1) - (A^J_{J+1})^2(2J+3) = a_J(1-a_J), \qquad (13)$$

the first of which is obtained by calculating the diagonal matrix element of the operator $\mathbf{Q}^2$, and the second—of the operator $Q^+Q^- - Q^-Q^+$. Solving the system (13), we obtain

$$A^J_{J+1} = \left[\frac{S(S+1) - a_J^2 J^2 - a_J J}{4(J+1)^2 - 1}\right]^{1/2},$$

$$A^J_{J-1} = \left[\frac{S(S+1) - a_J^2 J^2 + a_J[J(1-2a_J) + 1 - a_J]}{4J^2 - 1}\right]^{1/2}. \qquad (14)$$

We have thus determined the matrix elements (12), and with them also the Hamiltonian $\tilde{H}_1$ obtained above.

3. We now consider the indirect exchange induced by the Hamiltonian (8). In second-order perturbation theory we have

$$H_2 = \sum_{n\,m} J(\mathbf{R}_{nm})[{}^1\!/_2 N_n N_m + 2a_J^2 \mathbf{J}_n\mathbf{J}_m + 2a_J \mathbf{Q}_n\mathbf{J}_m]$$
$$+ 2\sum_{n,m} \mathcal{J}(\mathbf{R}_{nm})[a_J \mathbf{J}_n\mathbf{Q}_m + \mathbf{Q}_n\mathbf{Q}_m], \qquad (15)$$

$$J(\mathbf{R}_{nm}) = \sum_{\mathbf{k},\,\mathbf{k}'} |I_{\mathbf{k}\mathbf{k}'}|^2 \exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{R}_{nm}\}\frac{f_{\mathbf{k}}(1-f_{\mathbf{k}'})}{E_{\mathbf{k}} - E_{\mathbf{k}'}},$$

$$\mathcal{J}(\mathbf{R}_{nm}) = \sum_{\mathbf{k},\,\mathbf{k}'} |I_{\mathbf{k}\mathbf{k}'}|^2 \exp\{i(\mathbf{k}-\mathbf{k}')\mathbf{R}_{nm}\}\frac{f_{\mathbf{k}}(1-f_{\mathbf{k}'})}{E_{\mathbf{k}} - E_{\mathbf{k}'} - \Delta}, \qquad (16)$$

where $E_{\mathbf{k}}$ is the kinetic energy of the conduction electron, and $f_{\mathbf{k}}$ is the Fermi distribution function, which we assume here (in accordance with[11-13]) to be the same for both projections of the conduction-electron spin. If we discard the terms containing the operators $\mathbf{Q}$, then expression (15) goes over into the Hamiltonian H_0 of formula (31) from[5] 1). Thus (15) is a generalization of the indirect-exchange Hamiltonian to the case when the excitations of the total angular momentum of the f-shell are taken into account.

We see from (15) and (16) that, unlike expres-

sions (4) and (5) given in the paper of Mattis[9], the coefficients $2\alpha_J^2 J(\mathbf{R}_{nm})$ of the exchange integrals of the type $\mathbf{J}_n\mathbf{J}_m$ did not depend on the energy Δ. It follows therefore that the s-f exchange connected with the change in J cannot lead to the long-range ferromagnetic order discussed in[9]. Formally expressions (4) and (5) from[9] differ from our formulas (15) and (16) in the fact that in[9] the operators $\mathbf{Q}_n$ are replaced by $(Q_n^2)^{1/2}$, where the diagonal matrix element Q_n^2 of the operator $\mathbf{Q}_n^2$ is equal to

$$S_n(S_n+1) - a_{J_n}^2 J_n(J_n+1).$$

However, in[9] there are retained the operator products $\mathbf{J}_n\mathbf{J}_m$, which can not describe the change in J (since the operator $\mathbf{J}$ changes only the projection of the momentum, but does not change its magnitude). Actually, as seen from (15), the indirect exchange connected with the change in the total angular momentum of the f-shell is described by the operators Q. In the case when $n \neq m$, products of the type $\mathbf{Q}_n\mathbf{Q}_m$ can have nonvanishing diagonal matrix elements only for f-shells which are superpositions of the atomic wave functions with different J (in analogy with the "subantiferromagnetic" states, discussed by us earlier[10], which are superpositions of states with different S).

In the case when $n = m$, the operator Q_n^2 has the aforementioned diagonal element in a state with definite value of J_n, and accordingly the terms connected with the change in J (in the intermediate state) make a contribution

$$2\mathcal{J}(0)[S_n(S_n+1) - a_{J_n}^2 J_n(J_n+1)]$$

to the proper energy of the n-th f-shell, with spin S_n and total angular momentum J_n. We can therefore assume that the formula in the paper of Brout and Suhl[7] is of value only for the determination of the proper energy of the f-shell, but cannot be used to determine the magnetic order of the different f-shells.

Another important case, when the diagonal matrix elements of the operator Q_n^2 are of importance, is the interaction induced between the conduction electrons by the excitations J. This interaction will be considered in the next section.

4. It follows from (10) that the excitations J induce an interaction of singlet and triplet pairs of conduction electrons, given by the expression

$$H_{ee} = \frac{1}{\Delta}\sum_{\mathbf{k},\mathbf{k}',n}' \{G_{\mathbf{k}\mathbf{k}'} Q_n^2 B_{\mathbf{k}'}^{*S} B_{\mathbf{k}}^S + U_{\mathbf{k}\mathbf{k}'}[(2(Q_n^z)^2 - \mathbf{Q}_n^2)B_{\mathbf{k}'}^{*T}B_{\mathbf{k}}^T$$
$$- (Q_n^z)^2(B_{\mathbf{k}'\uparrow}^* B_{\mathbf{k}\uparrow} + B_{\mathbf{k}'\downarrow}^* B_{\mathbf{k}\downarrow})]\} \qquad (17)$$

where we have discarded the difference of the kinetic energies $E_{\mathbf{k}} - E_{\mathbf{k}'}$ compared with Δ; the

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1)We call attention to the omission from formula (31) of[5] of a factor n^2, which in our formula (15) corresponds to the product $N_n N_m$.

operators B_k^{*S}, $B_{k\uparrow}^{*T}$, $B_{k\uparrow}^*$, and $B_{k\downarrow}^*$, which we have introduced in[14], describe the creation of singlet and triplet pairs with spin projections S^Z respectively 0, +1, and −1; the prime in the double summation sign denotes that the summation extends over the half-space $k_z > 0$, $k_z' > 0$; the respectively even and odd matrix elements $G_{kk'}$ and $U_{kk'}$ are determined by the expressions

$$G_{k k'} = |I_{kk'}|^2 + |I_{-k,-k'}|^2 + |I_{-k,k'}|^2 + |I_{k,-k'}|^2,$$

$$U_{k k'} = |I_{kk'}|^2 + |I_{-k,-k'}|^2 - (|I_{-k,k'}|^2 + |I_{k,-k'}|^2). \quad (18)$$

The transitions of triplet pairs with $S^Z = 0$ and $S^Z = \pm 1$ enter in (17), generally speaking, with different coefficients. This difference disappears when all possible projections J^Z are encountered for the different impurity ions with equal probability, the value of J being specified for the ground state of the rare-earth ion. Indeed, using (12) and the first equation of (13) we can verify that the mean value of the diagonal elements of the operator $(Q^Z)^2$ is equal to the diagonal element of the operator $(1/3)\mathbf{Q}^2$. But this difference remains in force when the different values of J^Z turn out to have unequal probabilities. It follows therefore that the superconducting state realized by the triplet pairs can lift the degeneracy of the rare-earth in modulo J^Z (just as in the case of multiplicity excitation considered by us earlier[10]).

Let us assume here, however, that the superconductivity of lanthanum is realized by singlet pairs and that the character of these pairs does not change when a 1% rare-earth impurity is introduced. Then it follows from (17) that

$$H_{ee} = N_1 \Delta^{-1} \sum_{k,k_1}' G_{kk_1}[S(S+1) - a_J^2 J(J+1)]B_{k_1}^{*S}B_k^S,$$

$$\quad (19)$$

where N_1 is the number of impurities. The square bracket of (19) is equal to $(L+1)S/(J+1)$ in the case of the rare earths preceding Gd (for which $J = L - S$ in the ground state), and to LS/J for the remaining rare earths (for which $J = L + S$ in the ground state). In both cases the expression in the square bracket of (19) is positive and it follows therefore (taking into account the positiveness of $G_{kk'}$ in accordance with (18)), that the J excitations induce near the Fermi surface repulsion of singlet pairs of conduction electrons and, by the same token, a reduction in T_c. In particular, for Eu, with $J = 0$ and $L = S = 3$, we have

$$H_{ee} = 24 \frac{\zeta}{\Delta} \frac{I_0^2}{N} \sum_{k,k'} b_{k'}^* b_k = V_1 \sum_{k,k'} b_{k'}^* b_k, \quad (20)$$

where b^{*k} is the Bardeen-Cooper-Schrieffer[15]

pair-production operator, $\zeta = N_1/N$ is the impurity concentration, and $I_0 = NI$, where we replace $|I_{kk'}|$ near the Fermi surface by the constant value I.

For an estimate of V_1 we assume $\zeta = 1\%$[6], $\Delta \sim 1,000$ cm$^{-1} \approx 2 \times 10^{-13}$ erg[7], $N = 3 \times 10^{22}$[13], and $I_0 = 2.78 \times 10^{-13}$ erg according to[13] or $I_0 \approx 5 \times 10^{-13}$ erg according to[4]. We then obtain $V_1 \approx 4 \times 10^{-36} - 10^{-35}$. For pure lanthanum, the conduction-electron interaction parameter V can be estimated from the data for $N_0 V$ (N_0 is the density of the states near the Fermi surface), which is equal to 0.37[16], at $T_c = 5°$K and to 0.39[17] at the experimentally obtained[6] temperature $T_e = 5.7°$K. Assuming a conduction-electron concentration $n = 9 \times 10^{22}$[13], and accordingly a Fermi energy $E_F \sim 10^{-11}$ erg, we get $N_0 = 3n/4E_F \approx 6.7 \times 10^{33}$. We consequently obtain for the parameter V a value 6×10^{-35}, which is 15−6 times larger than the value of V_1 estimated above. Therefore, subtracting $V_1 - V$, we get approximately[2)]

$$T_c = T_{c0} \exp(-V_1 / N_0 V^2). \quad (21)$$

Substituting the foregoing estimates for V_1, N_0, and V and the value $T_{c0} = 5.7°$K for pure lanthanum, we find from (21) that 1% of Eu leads to $T_c \sim 4.8° - 3.8°$K. The second of these values of T_c coincides with the experimental value obtained by Matthias et al.[6]. Of course, one must not attach great significance to this agreement, in view of the approximate character of the estimates, but it is clear that this mechanism can provide, when reasonable estimates are used, not only a qualitative but also a quantitative explanation why T_c of a solution is lowered by ions that have no total angular momentum in the ground state.

It must be noted that the foregoing mechanism differs from the mechanism considered earlier[8,19,20] also in the character of the phase transition from the normal to the superconducting state in the presence of paramagnetic impurities. As is well known, the Herring[8] mechanism leads (see, for example, Sec. 4 in[20], dealing with the case when there is no ferromagnetism in either the normal or in the superconducting state) to a first-order phase transition, something not verified by experiment[21]. The mechanism which we consider, on the other hand, while changing the con-

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2)Strictly speaking, although the value of Δ given above exceeds by 15 times the Debye energy of lanthanum $\hbar\omega_D \approx 1.3 \times 10^{-14}$ erg, it would be necessary to subtract from V, taking into account the "logarithmic attenuation" of the repulsion[18], not V_1 but $V_1 [1 + N_0 V_1 \ln(\Delta/\hbar\omega_D)]^{-1}$. However, since in our estimates $N_0 V_1 \ln(\Delta/\hbar\omega_D) \sim 0.01 - 0.1$, we are justified in simply subtracting V_1 from V.

Ion	State	T_c, theor	T_c, exp	Ion	State	T_c, theor	T_c, exp
Pr^{3+}	$^3H_4$	5.4	5.25	Dy^{3+}	$^6H_{15/2}$	5.3	3.76
Nd^{3+}	$^4I_{9/4}$	5.3	4.7	Ho^{3+}	$^5I_8$	5.4	5.2
Sm^{3+}	$^6H_{5/2}$	4.9	4.5	Er^{3+}	$^4I_{15/2}$	5.5	5.3
Eu^{3+}	$^7F_0$	3.8	3.8	Yb^{3+}	$^2F_{7/2}$	5.6	5.5
Tb^{3+}	$^7F_6$	5.4	2.5				

duction-electron interaction parameter, does not violate the character of the phase transition as compared with pure La.

We note that a second-order phase transition is brought about also by the mechanism of conduction-electron scattering by paramagnetic impurities, considered by Abrikosov and Gor'kov[22]. They have assumed, however, that the good quantum number is S—the total spin of the f-shell, and not its total angular momentum. This raises the question of the degree to which the mechanism considered by us causes the T_c of lanthanum to be lowered by impurities other than Eu, impurities for which $J \neq 0$ in the ground state. Recognizing that for Eu the square bracket of (19) is equal to $S(S + 1)$, and writing for the critical temperature, in accord with (21),

$$T_c = T_{c0}e^{-\alpha S(S+1)}, \qquad (22)$$

we obtain $\alpha = 0.033$ for $T_c \approx 3.8°K$. Assuming as an estimate that I_0, Δ, and consequently also α are the same for all the rare-earth ions, we obtain accordingly for the critical temperature of La with 1% rare-earth impurities the expression

$$T_c = T_{c0}e^{-\alpha x}, \qquad (23)$$

where $x = (L + 1)S/(J + 1)$ or $x = LS/J$ for the ions situated to the left and to the right of Gd, respectively.

The values of T_c calculated in accordance with (23) are listed in the table[3], from which we see that the experimental values are systematically lower than the calculated ones (with the exception, of course, of the Eu impurity). It follows therefore that although the J excitations contribute to the lowering of T_c of lanthanum by all the rare earths, in the case of a ground state with $J \neq 0$ there is no doubt that other mechanisms, not connected with the change in J, play an important role.

One such mechanism is the scattering of the conduction electrons by paramagnetic impurities,

considered by Abrikosov and Gor'kov[22]. Indeed, if we assume that J is constant, make in formula (1) of[22] the substitution $S \rightarrow \alpha_J J$, and assume further in the calculations that

$$\overline{J^x} = \overline{J^y} = \overline{J^z} = 0, \qquad \overline{J^i J^k} = \frac{1}{3}J(J + 1)\delta_{ik},$$

then we find in accordance with (23) of[22] that at low impurity concentrations the decrease in T_c is proportional to $\zeta\alpha_J^2 J(J + 1)$. This dependence reduces (as shown in[19], where it was obtained on the basis of Herring's point of view[8]) to the form $\zeta S^2/(1 + 1/J)$ to the left of Gd and $\zeta S^2(1 + 1/J)$ to the right of it, which is in satisfactory agreement with the experimental data, except those for the Eu impurity.

Another mechanism that leads to a lowering of T_c of a superconductor with singlet pairs (possibly becoming significant at $\zeta \sim 1\%$) is inelastic scattering of the conduction electrons by systems of several paramagnetic ions [4], the simplest example of which are pairs of neighboring paramagnetic ions[23]. This question, however, is beyond the scope of the present article.

5. Thus, the s-f exchange Hamiltonian (10) obtained in this paper and connected with the changes in J, and also the indirect-exchange Hamiltonian (15) and the conduction-electron interaction Hamiltonian (17), which were derived on its basis, allow us to investigate the distinguishing features of the processes accompanied by excitation of the total angular momentum of the rare-earth ions. The main physical conclusions obtained for the processes considered above are essentially the following.

The excitations of J cannot lead to ferromagnetic ordering of different rare-earth ions. These excitations, however, can influence the occurrence of ordered states other than ferromagnetic, characterized by superposition of states with different J (for example, "subantiferromagnetic" order[10]).

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[3]The table does not include the elements Pm and Tm, for which there are no experimental data, Gd, for which L = 0, and Ce, which leads to an anomalous lowering of T_c, connected with the realignment of the electron configuration[6].

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[4]Such systems, in metallic systems Cu-Mn, were investigated experimentally, for example, by Znamenskii and Fakidov[24] at Mn concentrations $\sim$ 2.4, 5.3, 7.5, and 22.8 at.%. Electron-microscope data and a discussion of the stages of their production and growth in metallic systems subjected to quenching and aging can be found, for example, in[25].

In addition, these excitations of J can contribute to the proper energy of the rare-earth ions.

The interaction of the conduction electrons, induced by excitations of J, has a repulsion character for singlet pairs of conduction electrons. In the case of triplet pairs, this interaction is determined by an odd matrix element (which is consequently, generally speaking, of alternating sign).

Repulsion of singlet pairs of conduction electrons, induced by excitations of J, plays a major role in the case of the lowering of T_C of lanthanum by 1% Eu impurity, in which J = 0 in the ground state. In the case of admixture of other rare-earth ions, the principal role is apparently played by other mechanisms, not connected with the changes in J. It must be recognized here, however, that the good quantum number in this case is not the spin, but the total angular momentum J of the f-shell. An account of this circumstance, within the framework of the calculations given by Abrikosov and Gor'kov[22], leads to satisfactory agreement with experiment.

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Translated by J. G. Adashko
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