Electron spin resonance due to localized moments in superconducting metal oxides

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(Submitted 31 July 1987)
Zh. Eksp. Teor. Fiz. **94**, 276–284 (April 1988)

The ESR spectrum of Cu^{2+} ions in the superconducting $YBa_2Cu_3O_x$ system has been detected and identified. The most probable positions of localized Cu^{2+} moments in the structure of the compound $YBa_2Cu_3O_7$ have been established. The ESR spectrum of the $Y_{1-y}Gd_yBa_2Cu_3O_x$ system has been recorded for the first time. The temperature dependence of the linewidth of the normal phase has been used to find the integral of the exchange interaction between band and localized f electrons, amounting to $J_{sf} = 7.4 \times 10^{-3}$ eV. The depression of the superconducting transition temperature by localized moments of gadolinium in $GdBa_2Cu_3O_x$ has been estimated to be $\Delta T_c \approx -0.75$ K. At temperatures below T_c the ESR linewidth varies nonmonotonically with temperature. This can be explained by the appearance of a gap in the spectrum of single-particle excitations in the superconductor characterized by a parameter $2\Delta_0/kT_c \approx 6.5$. The integral representing the exchange interaction between Gd^{3+} ions has been estimated: $J(\text{Gd–Gd}) \approx 0.12$ K. The feasibility of magnetic ordering of gadolinium ions in the superconducting phase of $GdBa_2Cu_3O_x$ at temperatures below 7 K has been established.

1. INTRODUCTION

The discovery of superconductivity in the La–Ba–Cu–O system at temperatures above 30 K (Ref. 1) started an intensive search for new superconducting metal oxides. It was found very quickly that the Y–Ba–Cu–O system is superconducting at temperatures above 90 K (Ref. 2). In various papers reporting different investigations of magnetic properties of the Y–Ba–Cu–O system (see, for example, Ref. 3) that it has been noted the magnetic susceptibility exhibits a strong temperature dependence in the normal state, and this may be evidence of the presence of localized moments in the system. However, the available experimental results do not allow us to draw a definite conclusion about the nature of the paramagnetism of Y–Ba–Cu–O. Another interesting property of metal oxides of this type is that the replacement of yttrium with any rare-earth element has practically no effect on the superconducting transition temperature,[3] although one might expect magnetic states of rare-earth ions to be very different. This observation is not yet fully understood.

In investigations of the paramagnetism of solids the most detailed information on the nature of paramagnetic centers and on the microstructure of their environment is provided by the ESR method. In the case of localized magnetic moments in metals the ESR method also yields the magnitude of the integral representing the exchange interaction of conduction electrons with localized moments, which is responsible for depressing the superconducting transition temperature T_c by magnetic impurities. The temperature dependence of the rate of magnetic relaxation of localized magnetic moments below T_c can be used to judge changes in the spectrum of elementary excitations of a superconductor. Finally, the ESR method can be used to investigate directly the possibility of coexistence of a magnetically ordered state and superconductivity. These advantages of the ESR method have been utilized earlier in studies of type II superconductors doped with paramagnetic impurities (see, for example, Refs. 4–6).

We felt it would be of interest to use the ESR method to study the new class of metal-oxide superconductors with the structural formula $RBa_2Cu_3O_x$, where R is yttrium or any rare-earth element. Here we report the results of an ESR investigation of the $Y_{1-y}Gd_yBa_2Cu_3O_x$ system ($y = 0$, 0.02, 0.1, 1.0). These results are presented as follows. In the second section we describe briefly the investigated samples and the experimental method. In the third section we give the experimental results obtained from the ESR spectra of $YBa_2Cu_3O_x$, leading to the conclusion that the paramagnetism of these samples is due to localized moments of the Cu^{2+} ions. In the fourth section we give an account of an ESR investigation of the system $Y_{1-y}Gd_yBa_2Cu_3O_x$, ananalysis of which made it possible to determine the integral representing the exchange interaction between the localized f electrons of gadolinium and the band electrons, and to establish the possibility of magnetic ordering of the localized moments of gadolinium in the superconducting phase. The results obtained are discussed in the fifth section.

2. EXPERIMENTAL METHOD

Our samples were prepared by the now-familiar method from oxides of elements taken in the stoichiometric proportions corresponding to the formula $RBa_2Cu_3O_x$. An x-ray phase analysis showed that in addition to the main phase our $YBa_2Cu_3O_x$ samples contained a small amount (from 5 to 8%) of the "green" phase Y_2BaCuO_5. The temperature of the superconducting transition of all the samples was of the order of 90 K and the width of the transition determined by the inductive method was less than 2 K. Measurements of the upper critical field H_{c2} of $YBa_2Cu_3O_x$ in the temperature range 90–70 K gave the value $|dH_{c2}/dT| = 25$ kOe/K.

 0038-5646/88/040805-05$04.00

The ESR measurements were carried out using a BER-418[s] modulation spectrometer at a frequency of order 9400 MHz in the range 1.5–300 K. An increase of the signal/noise ratio was achieved by grinding the samples into a powder, which was mixed with molten paraffin and then cooled. Some ESR measurements were also made on oriented samples of $YBa_2Cu_3O_x$ and $Y_{1-y}Gd_yBa_2Cu_3O_x$. This was done as follows: a powder with a particle size on the order of individual grains (2–7 μm) was mixed with paraffin. Then an ampoule containing this mixture was subjected to a magnetic field $H = 10$–50 kOe. Paraffin was heated until completely softened and was then cooled in a magnetic field. An x-ray diffraction analysis showed that as a result of this procedure the crystallographic axes c of a large number of grains were oriented along the magnetic field direction.

3. ELECTRON SPIN RESONANCE OF $YBa_2Cu_3O_x$

The ESR spectrum of $YBa_2Cu_3O_x$ samples was recorded beginning from room temperature. Figure 1a shows an ESR spectrum of such a sample typical of temperatures above T_c. It is clear from this figure that the spectrum was of the kind usually obtained for powder samples containing ions with an axisymmetric anisotropic g factor. A satisfactory numerical simulation of this spectrum was possible only by adding a dispersion signal (about 20%) to the absorption signal, as usually observed for small conducting samples. It was found that $g_{\parallel} = 2.200 \pm 0.005$ and $g_{\perp} = 2.060 \pm 0.005$. These values of the g factor were typical of the ESR signals of the Cu^{2+} ions in an axial crystal field; the ground state of these ions can be described mainly by the wave function $d_{x^2-y^2}$, and not d_{z^2}, for which we would expect $g_{\perp} > 2.0023 \gtrsim g_{\parallel}$ (Ref. 7). In principle, we can postulate the appearance of localized Cu^{3+} moments for which the ground state is an orbital singlet with $S = 1$ (Ref. 7). However, a strong spin–orbit coupling results in a large peak-to-peak amplitude of the fine structure, so that observation of an ESR signal at room temperature would be unlikely for a powder sample.

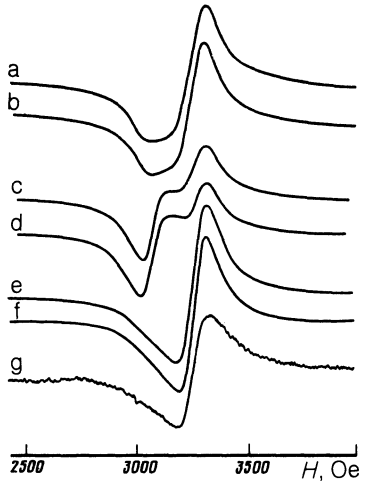

FIG. 1. Electron spin resonance spectra of $YBa_2Cu_3O_x$ in the normal state at $T = 130$ K: a) powder sample; c), e) oriented sample with the crystallographic c axis parallel and perpendicular to the direction of the resonance magnetic field, respectively; b), d), f) results of numerical simulation of the spectra of a powder and oriented samples, the latter for $c \| H_0$ and $c \perp H_0$, respectively. The simulation was carried out assuming the following parameters: $g_{\parallel} = 2.2$, $g_{\perp} = 2.06$, homogeneous line width $DH = 100$ Oe, oriented fraction 60%. The curve denoted by g is the ESR spectrum in the superconducting state at $T = 80$ K for $c \perp H_0$.

Since an x-ray phase analysis of our samples revealed the presence of the "green" phase, the features of the spectrum were identified by preparing a control sample of Y_2BaCuO_5. A comparison of the integrated ESR signals due to the same amounts of the pure green phase and of the investigated superconducting samples showed that the presence of 5–8% of the green phase could give rise to a signal approximately 15 times lower than that observed. An additional confirmation that the ESR signal of $YBa_2Cu_3O_x$ was different from that observed for the green phase was provided by an investigation of the evolution of the spectrum when temperature was varied. Cooling broadened the ESR signal of the superconducting sample and reduced the amplitude of this signal because broadening had the effect that a weak signal due to Y_2BaCuO_5 was observed superposed on this wide line. We shall now concentrate our attention on the spectrum of the main phase in a sample of $YBa_2Cu_3O_x$. Rough estimates indicated that $\sim 10\%$ of the copper ions present in this phase participated in the resonance, which made it possible to explain completely the temperature-dependent part of the magnetic susceptibility of the sample by the presence of the Cu^{2+} ions.

The positions of localized moments in the structure of $YBa_2Cu_3O_7$ were determined[8] by preparing an oriented sample in accordance with the method described above. The spectra obtained for two orientations of this sample relative to the resonance magnetic field H_0 are presented in Figs. 1c and 1d. In spite of the fact that the magnetic field oriented only partly the powder particles, we concluded from Figs. 1c and 1d that the symmetry axis of the local environment of the Cu^{2+} ions was oriented along the crystallographic c axis. This conclusion was confirmed by numerical simulation of the ESR spectra for these two orientations (Figs. 1d and 1f).

The paramagnetic Cu^{2+} centers are most probably in the planes between barium ions and these planes contain chains of copper and oxygen ions. This is supported by the instability of the oxygen ions in the plane in question, deduced from the results of x-ray structure analysis (see, for example, Ref. 9), and by our data on the correlation of the number of localized states with the heat treatment. Redistribution of oxygen ions in the xy plane gives rise to two types of local environment of copper ions. If oxygen atoms are "lost" in chains, the ground state of the Cu^{2+} ion should be d_{z^2}. The ESR spectrum of this state was not observed in our experiments. If oxygen atoms occupy vacant spaces between the chains, the Cu^{2+} ions are in a near-octahedral environment. According to the structure data of Ref. 9, the Cu–O distance along the $z \| c$ direction is less than the lengths of the Cu–O bonds in the xy plane. However, the inequality $g_{\parallel} > g_{\perp}$ we found experimentally corresponds to elongation of the octahedra along the c axis.[4)] The reason for the disagreement between the structure and spectroscopic data is not yet clear.

The temperature dependence of the line width and its position are given in Fig. 2. Note the absence of a significant Korringa contribution to the width of the ESR line increasing linearly with temperature at temperatures above T_c. It follows that the localized moments of Cu^{2+} are not in contact with the band electrons.

Small changes in the line width at ~ 150 K (this tem-

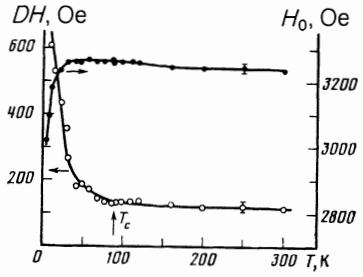

FIG. 2. Temperature dependencesof the resonance field and of the width of the ESR line of Cu^{2+} in an oriented sample of $YBa_2Cu_3O_x$ obtained for $c \perp H_0$.

perature ranges from 120 to 250 K, depending on the sample) were correlated with changes in the resonance field (g factor). This can be due to an electronic or a structural phase transition in a sample.

The transition of a sample to the superconducting state increased strongly the noise level (Fig. 1g), as we observed earlier for conventional superconductors[4] in which the noise was mainly due to the motion of an Abrikosov vortex lattice because of modulation of the external magnetic field. Usually the "zero" line reflecting the dependence of the nonresonance part of the surface impedance of a superconductor on the magnetic field behaved nonmonotonically, particularly in low magnetic fields where it exhibited a peak resembling resonance absorption. The integrated intensity of the ESR signal superimposed on this zero line was somewhat less than that in the normal state. The temperature dependence of the line width DH plotted on a double logarithmic scale revealed a kink in the region of 40 K, indicating two line-broadening mechanisms contributing to DH in different temperature intervals. We concluded that the line broadening at temperatures 90–40 K, when the resonance field was still unaffected, was due to the contribution made to the line width by an inhomogeneous broadening mechanism related to an Abrikosov vortex lattice. This contribution could be estimated roughly using the Ginzburg–Landau theory and the results of measurements of $H_{c1}(T)$ and $H_{c2}(T)$, from which the Ginzburg–Landau parameter $\varkappa \approx 70$–100 could be determined. Calculations similar to those reported in Refs. 4 and 5 indicated that for $\varkappa \gg 1$ the contribution to the width of the ESR line due to an inhomogeneous distribution of fields in the vortex lattice could be estimated from the expression

$$\Delta H_v \approx 0.1 H_{c2}^2/\varkappa^4 DH. \quad (1)$$

An estimate gave $\Delta H_v \sim 40$ Oe at $T = 40$ K, which was close to the observed broadening (Fig. 2). Therefore, the ESR signal of the Cu^{2+} ions behaved, on the one hand, as if the localized moments were located in the superconducting parts of the sample. In numerical simulation of the spectrum it was necessary to add, as an impurity, a dispersion signal to the absorption signal; the transition to the superconducting state increased strongly the noise level, reduced the signal intensity, resulted in a strong drift of the zero line, and made a contribution to the line width because of a vortex lattice. On the other hand, the results of measurements of the line width at temperatures above T_c indicated that the localized moments were not in contact with conduction electrons. These observations were insufficient to conclude that the parts of the sample where the ESR signal was observed were located spatially in the superconducting regions.

Broadening of the ESR signal below 40 K accompanied by a shift toward lower magnetic fields (Fig. 2), resembled the behavior of the ESR spectrum on approach to a magnetic ordering point. However, the considerable "width" of such a process on the temperature scale indicated a strong dispersion of the exchange fields, which was clearly associated with the spatial disorder of the localized Cu^{2+} states in the xy plane.

4. ELECTRON SPIN RESONANCE OF $Y_{1-y}Gd_yBa_2Cu_3O_x$

The ESR spectrum of a sample with $y = 0.02$ recorded at temperatures above T_c is shown in Fig. 3a. We observed a line of width $DH = 240$ Oe and $g \approx 2.0$, and we found that in weak magnetic fields an additional signal was observed when the spectrometer gain was high. The nature of this additional signal was investigated by applying an orienting magnetic field to the sample in the same way as in the case of a sample of $YBa_2Cu_2O_x$. The angular dependence of the spectrum of the oriented sample is demonstrated in Figs. 3b and 3c. In the $c \perp H_0$ direction the spectrum clearly shows an additional signal, whereas in the case of $c \| H_0$ the line becomes broader. We shall assume that this is due to the partly resolved fine structure of the ESR spectrum of Gd^{3+} ions. In the $c \perp H$ orientation (Fig. 3c) the splitting of the spectrum is 1500 Oe, which represents half the maximum peak-to-peak amplitude of the fine structure. This makes it possible to estimate the fine-structure constant of the ESR spectrum, which is $D \approx 0.02\ \text{cm}^{-1}$. In the $c \| H_0$ direction the total peak-to-peak amplitude of the fine structure is not observed and this is clearly associated with the orthorhombic ordering in the immediate environment of the Gd^{3+} ions. This ordering gives rise to an angular dependence of the spectrum in a plane perpendicular to the crystallographic c axis, and when a sample is oriented by a magnetic field, the disorder remains in this plane. Consequently, the fine structure is smeared out and cannot be observed.

Samples with $y > 0.02$ could not be oriented by a magnetic field. In the case of an unoriented sample with $y = 0.1$ we observed a spectrum similar to that shown in Fig. 3c.

The most interesting results we obtained were for a sample of $GdBa_2Cu_3O_x$. In this case we found that at tem-

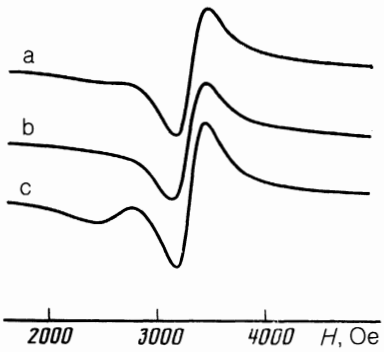

FIG. 3. Electron spin resonance spectrum of Gd^{3+} in $Y_{0.98}Gd_{0.02}Ba_2Cu_3O_x$ at $T = 150$ K: a) powder sample; b), c) oriented sample with $c \| H_0$ and $c \perp H_0$, respectively.

peratures above T_c there was a single smooth line with $g = 2.03 \pm 0.02$, which we identified as the ESR signal of the Gd^{3+} ions. The derivative of the absorption curve had the characteristic (of conducting objects) asymmetric form due to an admixture of the dispersion signal. The observed linear temperature dependence of the line width DH characterized by $d(DH)/dT = 0.82$ Oe/K (Fig. 4) was clearly due to the Korringa relaxation of the localized moments of gadolinium to conduction electrons:

$$DH_k = \frac{\pi k_B}{g\mu_B} (\rho_F J_{sf})^2 T. \qquad (2)$$

Here, ρ_F is the density of states of conduction electrons at the Fermi level and J_{sf} is the exchange integral of the interaction between the localized f electrons and conduction electrons.

A rough estimate of the density of states of conduction electrons at the Fermi level occurring in the above expression can be obtained if we know the electron specific heat $\gamma = 4.6 \times 10^{-3}$ J (mol Cu)$^{-1}$ K^{-2} obtained from our values of the upper critical field and of the electrical resistivity. We estimated that $\rho_F \approx 0.8$ eV^{-1} atom^{-1} spin^{-1}, enabling one to find $J_{sf} = 7.4 \times 10^{-3}$ eV from Eq. (2). Similar low values of J_{sf} were reported earlier[10] for rare-earth ions in molybdenum chalcogenides.

Using these values of J_{sf} and ρ_F, we could estimate the depression in the superconducting transition temperature[11] due to the presence of localized moments of gadolinium ions:

$$\Delta T_c = -\frac{\pi^2}{8k_B} cS(S+1)\rho_F J_{sf}^2. \qquad (3)$$

Here, S is the spin of localized states and c is the density of these states. In the case of $GdBa_2Cu_3O_x$ Eq. (3) gives $\Delta T = -0.75$ K. This is in good agreement with the experimentally observed weak dependence of T_c on the nature of the rare-earth ions in systems of this kind.

The temperature-independent residual line width, obtained by linear approximation of $DH(T)$ to $T = 0$ K, was found to be 720 Oe. The main contributions to the residual line width of a powder sample were made by the dipole–dipole interaction, the unresolved fine structure, and the anisotropy of the g factor. The last contribution could be ignored in the case of the Gd^{3+} ions in the S state. As pointed out above, the total peak-to-peak amplitude of the fine structure was of the order of 3000 Oe. A calculation of the dipole contribution to the line width gave a value of the order of 2500 Oe. Therefore, the observed residual line width amounting to 720 Oe was less than these contributions to DH, indicating that exchange narrowing of the ESR line took place. The exchange integral for the gadolinium ions can be estimated from an expression for the width of the ESR line in the method of moments:

$$DH = \pi (M_2^{3/2}/M_4^{1/2})\, /2\sqrt{3}. \qquad (4)$$

For this structure[8] and a given spin S, allowing only for the exchange interactions between the nearest neighbors and two coordination spheres for the dipole interactions, we can obtain the following expressions for the second and fourth moments:

$$M_2 = 2.5S(S+1)(g\mu_B)^4 a^{-6} + 15D^2,$$
$$M_4 = 7.7S(S+1)(g\mu_B)^4 a^{-6} J^2(\mathrm{Gd-Gd}). \qquad (5)$$

Using $S = 7/2$ and $a \approx 3.8$ Å, as well as our measured values $DH(0) = 720$ Oe and $D \approx 0.02$ cm^{-1}, we find that Eqs. (4) and (5) yield $J(\mathrm{Gd\text{–}Gd}) \approx 0.12$ K. The paramagnetic Curie temperature

$$\Theta_p = S(S+1)J(\mathrm{Gd-Gd})z/3$$

for $z = 4$ nearest neighbors is 2.4 K, which is comparable with the value $\Theta_p = -5$ K we used to describe the temperature dependence of the magnetic susceptibility of the same sample.

The exchange integral J_{sf} found with the aid of Eq. (2) makes it possible to estimate, in principle, the integral of the RKKY interaction of the magnetic Gd^{3+} ions via conduction electrons. In the free-electron approximation this estimate gives a value of the exchange integral $J(\mathrm{Gd\text{–}Gd})$ an order of magnitude less than that obtained above. This evidently demonstrates the important role of the superexchange mechanisms and of the details of the structure of the conduction band in the exchange interaction between the Gd^{3+} ions.

After some broadening near the superconducting transition temperature the ESR line decreases in width by about 75 Oe in the temperature range 80–50 K (Fig. 4). It is then found that the inhomogeneous contribution to the line width due to the vortex lattice, estimated on the basis of Eq. (1) ($\Delta H_v \approx 7$ Oe at $T = 50$ K) can be ignored. The behavior of DH resembles qualitatively the temperature dependence of the line width in the case of conventional superconductors (see, for example, Ref. 6) for which such a nonmonotonic behavior is associated with the presence of a gap in the spectrum of single-particle excitations. The appearance of a gap is accompanied by an increase in the density of electron states and by coherence effects which broaden the line directly below T_c. Further cooling reduces the line width because of the exponential reduction in the number of single-particle excitations. Attempts to describe the temperature dependence of the line width by means of the BCS theory were unsuccessful.

At temperatures below 50 K the ESR line begins to widen and it shifts toward lower values of the magnetic field (Fig. 4). This is clearly an indication of the freezing of the exchange fluctuations and loss of the exchange narrowing associated with the approach to the magnetic ordering point. Since the ESR line becomes undetectable below 7 K, without

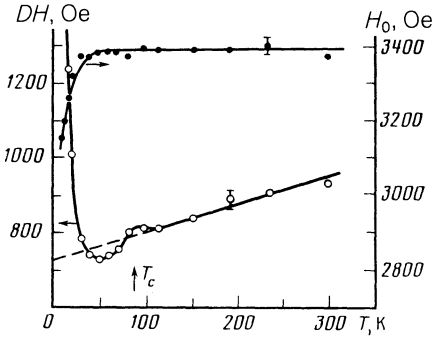

FIG. 4. Temperature dependences of the resonance field on the width of the ESR line of Gd^{3+} in $GdBa_2Cu_3O_x$. The arrow identifies the position of T_c, measured by the inductive method.

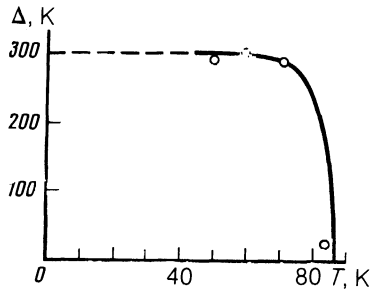

FIG. 5. Behavior of the energy gap deduced from the temperature dependence of the ESR line width for $GdBa_2Cu_3O_x$.

reaching the maximum value of the width, we can expect the magnetic ordering temperature to be $T_m < 7$ K. This agrees with the results of Ref. 12, in which the specific heat of $GdBa_2Cu_3O_x$ is used to draw the conclusion that $T_m = 2.24$ K.

5. CONCLUDING COMMENTS

In the previous sections we reported results of investigations of electron spin resonance in the metal oxide system $Y_{1-y}Gd_yBa_2Cu_3O_x$. The results we obtained from the ESR spectra of the Cu^{2+} ions suggest disorder of a considerable number of oxygen vacancies in copper–oxygen planes between barium ions, compared with the ideal structure of $YBa_2Cu_3O_x$ (Ref. 8). Bearing in mind that about one-third of the copper ions in these planes have localized moments, we may conclude that one-dimensional chains of copper and oxygen atoms are disturbed in large parts of our samples.

We are assuming that the immediate environment of the localized moments of the Cu^{2+} ions is nearly octahedral in the system under discussion. It is well known (see, for example, Ref. 5) that such complexes tend to exhibit distortions due to the Jahn–Teller effect which becomes cooperative in the presence of an interaction via the phonon field. An estimate of the interaction energy of localized d electrons of copper via the phonon field over distances of 8 Å (which corresponds to the average distances in our samples) gives 180 K (Ref. 13). Therefore, the observed small changes in the width and position of the resonance line at temperatures ~ 150 K (Fig. 2) may possibly represent a manifestation of the cooperative Jahn–Teller effect.

The temperature dependence of the ESR line width of the gadolinium ions above T_c gives the integral of the exchange interaction J_{sf} between the localized f electrons of gadolinium and conduction electrons. Assuming the most probable p- and d-nature of the wave functions of the band electrons on the Fermi surface, we are not surprised by the small value of J_{sf} or by the resultant weak depression of superconductivity by magnetic rare-earth ions.

The temperature dependence of the ESR line width of the Gd^{3+} ions below T_c demonstrates that the superconductivity appears in copper–oxygen planes adjacent to the gadolinium ions. The smallness of the Korringa contribution to the width of the line near T_c causes changes in the line width to exceed the experimental error only slightly. In spite of this, we can attempt to estimate the gap Δ in the spectrum of elementary excitations ensuring the observed temperature dependence of the ESR line width below T_c. We can do this using an expression for the rate of relaxation of the transverse component of the magnetization of the localized moments in a superconductor.[14] This gives the temperature dependence of the gap $\Delta(T)$, shown in Fig. 5. It is found that the gap near T_c rises rapidly, already reaching a constant value $2\Delta_0/k_BT_c \approx 6.5$ at $T \sim 70$ K. This may be evidence of the strong-binding superconductivity.

Our data are insufficient to determine the nature of the magnetic order of the gadolinium ions which appears at $T < 7$ K. Even the absence of "reentrant" superconductivity for the values of J_{sf} obtained by us does not justify exclusion of ferromagnetism. However, the negative sign of Θ_p suggests that antiferromagnetism is more likely.

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[4)]It should be pointed out that the relationship $g_{\parallel} > g_{\perp}$ may, in principle, be also realized in compressed octahedra if they are pairwise mutually perpendicular in the xy plane and the Cu^{2+} ions are coupled by the exchange. However, this seems unlikely.

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Translated by A. Tybulewicz