

Electron paramagnetic resonance in superconducting $\text{La}_{3-x}\text{Gd}_x\text{In}$

N. E. Alekseevskii,¹⁾ I. A. Garifullin, B. I. Kochalev,²⁾ G. G. Khaliullin, and É. G. Kharakhash'yan

Kazan' Physicotechnical Institute, Kazan' Branch, Academy of Sciences of the USSR

(Submitted 14 October 1981)

Zh. Eksp. Teor. Fiz. **82**, 1979–1989 (June 1982)

The magnetic state of gadolinium impurities in superconducting $\text{La}_{3-x}\text{Gd}_x\text{In}$ is investigated experimentally for values of x in the range 0.005–0.12. It is concluded on the basis of EPR, heat-capacity, and magnetic-susceptibility data that antiferromagnetic clusters arise at temperatures below 15 K in a specimen with $x = 0.12$, in which superconductivity is completely suppressed by the magnetic impurities, and that a magnetically ordered state with a small spontaneous moment appears in the same specimen at temperatures $T \lesssim 2$ K. The experimental results can be described with the aid of an exchange-interaction potential between the localized moments that are of antiferromagnetic type at distances of the order of the lattice constant and predominantly ferromagnetic at large distances. EPR studies of specimens with $x > 0.005$ revealed a broadening of the resonance line associated with magnetic ordering of the gadolinium impurities in the superconducting phase itself. An analysis of the experimental data indicates that magnetic ordering processes apparently take place in the superconducting phase in the same way as they do in the normal state.

PACS numbers: 74.70.Gj, 76.30.Kg, 75.30.Hx

After the publication of the first paper in which the presence of electron paramagnetic resonance (EPR) in a type-II semiconductor was reported¹ there appeared a large number of papers (e.g., Refs. 2–5) in which the EPR of gadolinium ions in the intermetallic compounds LaRu_2 , CeRu_2 , and ThRu_2 , which are superconductors with fairly high critical parameters, was investigated. The detailed analyses of the temperature dependences of the line width and g factor given in these papers made it possible to obtain information on the behavior of the electronic susceptibility of superconductors and on features of their electronic structure. Specimens containing only a small quantity of the magnetic impurity were used in these investigations to avoid broadening of the resonance line as a result of the spin-spin interactions. In addition, it was shown^{6,7} that EPR provides a most effective method for investigating the nature and strength of the spin-spin interactions between localized electronic states in the superconducting phase. It accordingly becomes possible to use the EPR method to investigate one of the most controversial problems in the physics of superconductors—that of the coexistence of magnetic order and superconductivity. A great many papers on the experimental study of this problem using various physical methods have been published (see the reviews by Maple⁸ and Roth⁹).

In the work reported here we used the EPR method for the first time³⁾ to investigate the effect of magnetic ordering of impurities in a superconductor.

The superconductive compound La_3In doped with Gd was chosen as the material for the investigation. This system is one of the superconductors with tight binding and is a very convenient superconductive matrix for investigating effects associated with the presence of magnetic impurities, since La_3In is a good solvent for rare-earth metals. Earlier studies^{11,12} of the dependences of the critical field and critical temperature on the gadolinium concentration in this system made it reasonable to suppose that superconductivity and magnetic order might coexist in $\text{La}_{3-x}\text{Gd}_x\text{In}$ ($x = 0.04$ cor-

responds to 1 at.% of Gd) at relatively high temperatures.

In the first part of the paper we present and discuss the data on the normal and superconductive properties of the specimens (measurements of the electrical conductivity, the superconducting transition temperature, the upper critical field, the susceptibility, and the heat capacity). The results of the EPR measurements are presented and discussed in the second part.

1. NORMAL AND SUPERCONDUCTIVE PROPERTIES OF THE SPECIMENS

Specimen preparation. The specimens were synthesized in an induction furnace in an atmosphere of pure helium from lanthanum, gadolinium, and indium of purity 99.88, 99.8, and 99.99 wt. %, respectively, in niobium crucibles. In view of the high chemical activity of the compound, the specimens for EPR measurements were flushed in the liquid state in containers of highly pure molybdenum that does not exhibit EPR. The resonance signal was recorded only from the chamfered face of the specimen, which was immediately shielded before the measurements. This avoided the presence of “parasitic” signals in the EPR spectrum from the oxide layer that would rapidly form in the air. Metallographic analysis of the finished specimens revealed the presence of two contaminant phases. Using the data of Ref. 13, we identified a phase of dendritic form as La_5In_3 , and the disseminations of the second contaminant phase, as La. At the same time, x-ray phase analysis of the specimens revealed only one phase having a Cu_3Au -type lattice with the lattice constant $a = 5.07$ Å, which corresponds to La_3In . Moreover, it was found that deviations in either direction from the stoichiometric proportions in preparing the specimens resulted in the appearance of additional reflections in the x-ray diffraction pattern. On the basis of these studies, we conclude that the upper limit of the bulk content of extraneous phases does not exceed 3–4%.

Electrical conductivity. The resistivity of the specimens at room temperature was $\rho(300\text{ K}) = 110 \times 10^{-6}$ ohm·cm, and the ratio of the resistivity at room temperature to that at the superconducting transition point decreased with increasing Gd content from 9.5 for La_3In to 8.0 for $\text{La}_{2.95}\text{Gd}_{0.05}\text{In}$.

Superconducting transition temperature and the upper critical field strength. The superconducting transition temperature T_c was determined from the temperature dependence of the inductance at 100 kHz of a coil surrounding the specimen. We found $T_c = 9.2\text{ K}$ for pure La_3In . The superconducting transition temperature T_c was found to decrease linearly with increasing gadolinium concentration c clear up to the concentration $c = 1.25\text{ at.}\%$, the slope being $dT_c/dc = -3\text{ K/at.}\%$. For specimens with low gadolinium concentrations ($c < 0.7\text{ at.}\%$) the width of the superconducting transition was smaller than 0.2 K; this indicates that the synthesized specimens were of satisfactory quality and that the impurity was distributed uniformly within them. It should be noted that the T_c values obtained from dc resistance measurements agree well with those obtained from the inductance measurements.

The upper critical field strength H_{c2} was determined from the change in the dc resistance.⁴⁾ Figure 1 shows the results of these measurements for several specimens; they are in good agreement with the results of other studies.^{11,12}

Susceptibility. The susceptibility measurements were made by the Faraday method in magnetic fields weaker than 400 Oe. Figure 2 shows the results of the magnetic susceptibility measurements on a $\text{La}_{2.95}\text{Gd}_{0.12}\text{In}$ specimen in which the superconductivity was fully suppressed by the gadolinium. The appearance of a small spontaneous magnetic moment in this specimen at 1.9 K, which may be taken as the magnetic ordering point T_M , was determined by the ballistic method.

As is evident from the figure, the susceptibility follows the Curie-Weiss law in the region $T > 15\text{ K}$. The considerable deviations from the Curie-Weiss law in the temperature interval $T_M < T \leq 15\text{ K}$ indicates the presence in this specimen of strong spatial dispersion of the exchange fields. The properties of such systems at temperatures $T \gg T_M$ can be investigated by the vir-

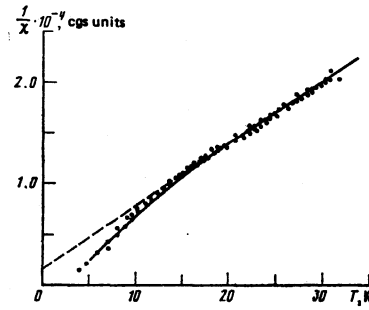


FIG. 2. Temperature dependence of the magnetic susceptibility in a 400-Oe magnetic field of a specimen containing 3 at. % of cadmium. The full curve was calculated.

ial-expansion method.¹⁴ In the pair-correlation approximation, the susceptibility in a weak magnetic field may be expressed in the form

$$\chi = \chi_0 \left[1 + c \sum_j f(J(R)/T) \right],$$

$$f(x) = \frac{1}{Z} \sum_{j=0}^{\infty} (2j+1) \left[\frac{\alpha_j}{S(S+1)} - 1 \right] \exp(\alpha_j x), \quad (1)$$

$$Z = \sum_j (2j+1) \exp(\alpha_j x), \quad \alpha_j = j(j+1)/2,$$

where χ_0 is the Curie susceptibility, c is the impurity concentration in atomic fractions, $J(R)$ is the strength of the exchange interaction between a pair of spins separated by the distance R , and S is the spin of the impurity. Numerical calculations show that if the Ruderman-Kittel approximation $J(R) = V_0 \cos(2k_f R)/R^3$ is adopted for the exchange integral, the observed dependence of χ on T cannot be reproduced by the expression (1) for any reasonable values of the coefficient V_0 and the Fermi momentum k_f .⁵⁾ The Ruderman-Kittel potential is evidently too crude an approximation for the compound under investigation. Actually, because of the strong temperature dependence of the susceptibility and the Knight shift in La_3In ,^{15,16} we may assume that there is a narrow peak in the density of states near the Fermi level. For this case a potential for the indirect exchange interaction through the conduction band has been found,¹⁷ which has the form

$$J(R) \approx \begin{cases} J_0 (\sin(k_f R)/k_f R)^2, & R \leq R_0 \approx 2\epsilon_f/k_f D \\ V_0 \cos(2k_f R)/R^3, & R > R_0 \end{cases}, \quad (2)$$

where D is the width of the peak in the density of states and ϵ_f is the Fermi energy. The coefficient J_0 is positive, so the interaction (2) favors parallel orientation of the spins out to distances R of the order of $R_0 \gg a$ and cannot account for the observed negative sign of the paramagnetic Curie temperature of the investigated specimen (Fig. 2). In addition to Eq. (2), therefore, we postulate that there is a strong antiferromagnetic interaction at close distances $R \sim a$. A possible reason for such interaction might be indirect exchange via nonmagnetic ions. The results of a calculation of expression (1) with $J(R) = -3\text{ K}$ for the first two coordination spheres and as given by Eq. (2) with $J_0 = 13\text{ K}$, $(k_f a) = 7$, and $2\epsilon_f/D = 23$ for $R \geq \sqrt{(\frac{3}{2})} a$ are shown in Fig. 2. We note that the agreement with the experimental suscep-

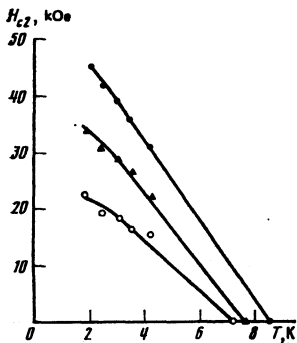


FIG. 1. Temperature dependence of the upper critical field for specimens containing 0.25 (●), 0.5 (▲), and 0.75 (○) at. % of gadolinium.

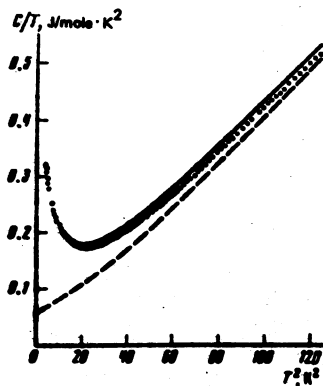


FIG. 3. Temperature dependence of the heat capacity of a specimen containing 3 at. % of gadolinium: the points represent our experimental data and the full curve was calculated. The dashed curve represents the experimental data of Ref. 15 for pure La_3In .

tibility data could be improved by varying these parameter values somewhat, but the parameter values given above were chosen so as to give the best description of all our experimental results, including the heat-capacity and EPR measurements.

Generally speaking, the specific form of the function $J(R)$ is not of critical importance; what is important is only that it assume large negative values for $R_1 \sim a$ and comparatively small positive values for $R > R_1$.

Heat capacity. The heat-capacity measurements were made using the setup described in Ref. 18. Figure 3 shows the results of such measurements on the $\text{La}_{2.98}\text{Gd}_{0.12}\text{In}$ specimen in the temperature range 1.95–13 K. Figure 3 also shows the data of Ref. 15 for “pure” La_3In , which were obtained in a 90-kOe magnetic field in order to fully suppress the superconductivity of the specimen.

Comparison with the heat capacity of pure La_3In shows that there is a weakly temperature dependent magnetic contribution C_M to the heat capacity at temperatures $T \geq 5$ K. This again indicates the presence in the crystal of strongly coupled antiferromagnetic pairs, as was assumed above. The weak long-range exchange forces described by the potential (2) are responsible for the increase in the magnetic contribution at temperatures $T < 5$ K.

The first term in the virial expansion of the heat capacity has the form

$$C_M(T) = \frac{1}{2} N k_B \sum_j U_j(T) F(U_j(T)) \quad (3)$$

$$F(x) = \frac{1}{2} \sum_j (2j+1) \alpha_j^2 \exp(\alpha_j x) - \left\{ \frac{1}{2} \sum_j (2j+1) \alpha_j \exp(\alpha_j x) \right\}^2$$

where N is the number of impurity atoms and k_B is Boltzmann's constant. The results of a numerical calculation of expression (3) for the values of the exchange integrals used to describe the magnetic susceptibility are presented in Fig. 3. It should be noted that the approximation (3) is useful only at high temperatures. The magnetic contribution to the heat capacity exhibits

a $(T - T_M)^{-\alpha}$ temperature dependence with $\alpha \approx 0.1$ in the region $1.95 \text{ K} < T < 3 \text{ K}$.

It is of interest to estimate the electronic heat capacity γ from the results of the residual-resistance and upper-critical-field measurements, using the well-known Gor'kov equation,¹⁹ which is valid for “dirty” superconducting alloys:

$$\gamma = 2.2 \cdot 10^{-3} \rho^{-1} dH_{c2}/dT. \quad (4)$$

Such an estimate for the specimen containing 0.75 at. % Gd ($\rho(T_0) = 12 \times 10^{-9}$ ohm·cm and $dH_{c2}/dT = 4.45 \times 10^3$ Oe/K) yields $\gamma = 64$ mJ/mole·K², which is in fairly good agreement with the value obtained in Ref. 15.

2. RESULTS AND DISCUSSION OF THE EPR MEASUREMENTS

The EPR measurements were made at temperatures from 1.6 to 50 K, using an HB-ER418S modulation spectrometer working in the 3-cm wavelength range. The temperature of the specimen during the measurements was determined, at temperatures below 4.2 K, by the helium vapor pressure, and at higher temperatures, with a carbon resistance thermometer.

For all the investigated specimens in the normal state, the profile of the EPR line from resonance at localized gadolinium states had the asymmetric form usual for massive metallic specimens. The g factor and the line width ΔH were determined using the technique developed in Ref. 20. The width of the EPR line in the superconducting state was determined with allowance for the change in the line profile as a result of the appearance of eddies.⁷ The results of the measurements of the resonance line width for the investigated specimens are presented in Figs. 4, 5, and 6.

The normal state. The temperature dependence of the line width in the normal state at high temperatures can be represented by a linear function of the form ΔH

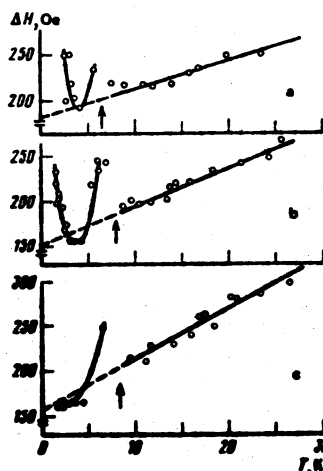


FIG. 4. Temperature dependence of the line width for specimens containing gadolinium at concentrations of 0.75 (a), 0.25 (b), and 0.125 (c) at. %. The arrows mark the transition temperature to the superconducting state in the resonance field.

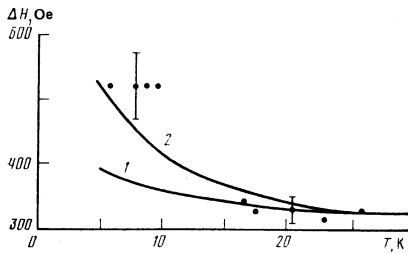


FIG. 5. Temperature dependence of the line width of a specimen containing 3 at.% of gadolinium. Curves 1 and 2 were calculated with formulas (8) and (9), respectively.

$= a + bT$ in which the coefficients a and b depend on the Gd concentration (see Table I). That the temperature gradient b of the line width depends on the Gd concentration indicates that the conditions for an electron bottleneck in the relaxation of localized moments are satisfied. In this case one can determine important dynamic characteristics of the conduction electrons from EPR data. The temperature gradient of the EPR line width is given by an expression of the form²¹:

$$b = \frac{\pi k_B J_{sf}^2 N_0^2}{g \mu_B} \frac{\delta_{eL}}{\delta_{ei} + \delta_{eL}}, \quad (5)$$

where μ_B is the Bohr magneton, g is the spectroscopic g factor, N_0 is the density of conduction-electron states at the Fermi surface, J_{sf} is the exchange-interaction integral for the interaction of conduction electrons with localized moments, δ_{ei} is the exchange-scattering rate, and δ_{eL} is the spin-lattice relaxation rate for the conduction electrons. Using the value $N_0 = 1.7$ (eV · atom · spin)⁻¹ that we determined from the dH_{c2}/dT data with the aid of the Gor'kov relation, we obtain $\delta_{eL} = (8 \times 10^{10})c$ sec⁻¹, $\delta_{ei} = (1.7 \times 10^{13})c$ sec⁻¹, and $J_{sf} = 0.01$ eV, where c is the gadolinium concentration in atomic percent.⁶⁾

One can also evaluate δ_{ei} from the dependence of the critical temperature of the superconductor on the magnetic-impurity concentration, using the well-known formula of Abrikosov and Gor'kov²²: $\Delta T_c = 3\pi\hbar \delta_{ei} / 16k_B$. The experimental value $\Delta T_c/c = 3$ K yields $\delta_{ei} = (6.7 \times 10^{13})c$ sec⁻¹. The difference between the values obtained for the exchange scattering rate from the EPR data and from the superconducting-transition-temperature data is evidently to be attributed to the possibility

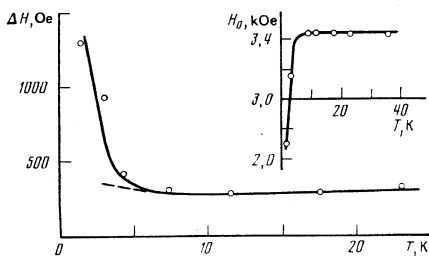


FIG. 6. Temperature dependences of the line width ΔH and the resonance value H_0 of the magnetic field for a specimen with a gadolinium concentration of 1.25 at.%. The dashed curve was calculated with formula (9).

TABLE I.

Gadolinium concentration, at.%	a , Oe	b , Oe/K	g -factor
0.125	157±8	5.6±0.2	1.98±0.01
0.25	152±8	4.2±0.2	1.98±0.01
0.75	182±9	3.0±0.2	1.98±0.01
1.25	220±15	2.0±0.2	1.95±0.02
1.5	260±20	<0.5	1.98±0.02
2.0	240±20	<0.5	1.98±0.02
3.0	320±20	<0.5	1.99±0.02

that nonmagnetic scattering of conduction electrons by gadolinium impurities may contribute appreciably to the breakdown of the superconductivity. In that case the estimate of the exchange-scattering rate based on the concentration dependence of the superconducting transition temperature would be too high. Further, the possibility cannot be entirely ruled out that the relaxation of localized gadolinium moments in La_3In and the superconductivity of that compound may be due to electrons of different bands. Moreover, it must be borne in mind that the electron bottleneck effect arises only when the relaxation of localized moments is due to s -band electrons, since d electrons have very short spin relaxation times.

The "residual" line width a in the metal is determined by the spin-spin interactions, the fine structure, and the inhomogeneities of the crystal lattice. Estimates show that even in the absence of exchange narrowing, the contribution of dipole-dipole interactions to the line width does not exceed 100 Oe at the concentrations used in the present work. For the investigated specimens, therefore, the line width is evidently determined mainly by the fine structure of the Gd^{3+} ion and the inhomogeneities of the crystal, whose contributions are partially averaged by spin fluctuations induced by indirect exchange interactions.

The contribution to the frequency of the spin fluctuations from the antiferromagnetic exchange pairs, which were mentioned above, freezes out at temperatures $T \lesssim 15$ K; consequently, the resonance line broadens with decreasing temperature, as is evident from Fig. 5. Then the temperature dependence of the line width is determined by the behavior of the correlation time

$$\tau_c(T) = \int_0^\infty dt g(t) \quad (6)$$

where $g(t) = \langle S_i^z(t) S_i^z \rangle / \langle S_i^z S_i^z \rangle$, the average being taken over the positions of the magnetic impurities. An estimate by the method of moments in the pair correlations approximation leads to the formula

$$g(t) = \prod_{k \neq i, p} \exp \left\{ -\frac{S(S+1)}{3} \left[1 + \frac{S_k S_p}{S(S+1)} J^2(R_{ik}) \right] \right\}, \quad (7)$$

in which S_p is the spin closest to S_k . The final result depends strongly on the order in which expression (7) is averaged over the spin configuration. If the argument of the exponential in (7) is first averaged over the distances R_{ik} to the nearest neighbor and then a statistical average is taken over the index k we obtain

$$\tau(T) = \tau_\infty \left\{ \sum_{\mathbf{R}} p(\mathbf{R}) (1 + f(J(\mathbf{R})/T))^{\mathbf{R}} \right\}^{-1}, \quad (8)$$

where τ_∞ is the limit of the correlation time as $T \rightarrow \infty$, $p(R)$ is the probability that the nearest spin will be at the point R , and the function $f(x)$ is defined in Eq. (1). But if we take the averages in the opposite order, we obtain a multiexponential correlation function with

$$\tau(T) = \tau_\infty \sum_i p(R) \{1 + f(J(R)/T)\}^{-1}. \quad (9)$$

Of course both these expressions must be regarded as only rough approximations, since formula (9) strongly overestimates the contribution from configurations in which the i -th spin is surrounded by bound pairs and the frequency of its fluctuations is low, whereas formula (8) does not take that situation into account at all. Figure 5 shows plots of ΔH vs T as given by the formula $\Delta H = a\tau(T)/\tau_\infty + bT$ with a and b taken from Table I and the function $\tau(T)$ calculated from formulas (8) and (9), using the values of the exchange integrals found above. As the figure shows, the second limiting case gives the better agreement with experiment.

At low magnetic-impurity concentrations when there are few antiferromagnetic pairs, the line-broadening mechanism discussed above becomes less effective and is almost entirely cancelled out by the temperature dependence of the line width given by Eq. (5) (see Fig. 6).

As the temperature approaches the magnetic ordering point the correlation range increases sharply under the action of the exchange potential (2), and this leads to a corresponding increase in the line width and to a shift of the resonance toward the weaker magnetic fields. In the normal state, this effect is most clearly seen in the specimen having the 1.25-at.% Gd concentration (Fig. 6).

On the basis of all the experimental data, we can sketch the following qualitative picture of the magnetic state of the impurities in the investigated compound. At temperatures $T \lesssim 15$ K there is established in the specimen a strong antiferromagnetic correlation between spins lying close together. As the temperature decreases further, the weak exchange interactions, preferentially of ferromagnetic type at distances $R \lesssim 3\text{\AA}$, lead to "freezing" of the spin system with the formation of a complex magnetic structure with a small spontaneous moment.

It must be noted that in their experimental study of the magnetic properties of the $\text{La}_{3-x}\text{Gd}_x\text{In}$ system the authors of Ref. 23 found considerably larger deviations from the Curie-Weiss law than we did; in our opinion, this difference is due to the difference between the methods used in the two studies for preparing the specimens. In discussing the gigantic values of the magnetic susceptibility reported in Ref. 23, the authors of Ref. 24 concluded that the gadolinium impurity was not randomly distributed throughout the alloy, but formed large quasi-ferromagnetic clusters when $T \gg T_M$. On the other hand, the temperature independence of the g -factor shift when $T \gg T_M$ and the fact that the intensity of EPR signal deviated only slightly from the Curie-Weiss law evidently give us reason to assume that the impurity was uniformly distributed in our specimens. Thus, the difference between our results and those of Ref. 23 once more emphasizes the sensitivity of the properties of

dilute magnetic alloys to the technique used in preparing the specimens.

The superconducting state. The specimens with gadolinium concentrations of 0.125, 0.25, and 0.75 at.% become superconductive at temperatures in the range 6–8 K, the EPR line width first increasing and then rapidly decreasing (Fig. 4). This behavior of the line width is analogous to the change in the nuclear relaxation rate in a superconductor and is associated with the corresponding change in the density of states and the appearance of a gap in the elementary excitation spectrum. As the temperature decreases below 3 K the line width for the specimen with the 0.125-at.% Gd concentration does not change further, while those for the specimens with the 0.25- and 0.75-at.% Gd concentrations increase again.

As was noted above, for specimens with low magnetic-impurity concentrations the line-broadening mechanism becomes largely ineffective at low temperatures because the contribution from antiferromagnetic pairs to the spin-fluctuation frequency freezes out. It is therefore natural to assume that the observed behavior of the line width for the specimens with Gd concentrations of 0.25 and 0.75 at.% at temperatures below 3 K is due to an increase in the range of the magnetic correlations under the action of long-range exchange interactions.

In investigating the coexistence of magnetic order and superconductivity it is important to answer the following question: Do superconductivity and magnetic order coexist at the same place, or are the superconductive and magnetic phases spatially separated? Since the basis for the conclusion that the gadolinium impurities are magnetically ordered in the superconducting phase is the characteristic behavior of the resonance line near the magnetic ordering point, it is necessary to analyze the possibility that a parasitic signal from extraneous inclusions in the normal state may be superimposed on the EPR spectrum. The normal phase might be produced either by a nonuniform distribution of the impurity throughout the specimen or as a result of precipitation of a nonsuperconducting modification of the investigated compound.

The degree of spatial uniformity of the impurity distribution can be estimated from measurements of T_c made by the induction method. This method provides information on the variance of T_c throughout the volume of the specimen, whereas measurements of T_c based on conductivity methods do not, since in principle it is possible that normal-state inclusions may be shorted out by superconducting regions having high critical temperatures. If it is assumed that the fluctuation of the Gd concentration in the specimen is the main reason for the broadening of the superconducting transition, the temperature dependences obtained for the volume that has passed into the superconducting state can be very well described under the assumption that the impurity is distributed normally in the specimen with a quite small standard deviation—for example, with a standard deviation of $\sigma_c = 0.08\%$ for a specimen with $c = 1.25\%$ (i.e., $1.05\% \leq c \leq 1.45\%$ throughout 98% of the volume

of the specimen). Moreover, as was noted above, the EPR and paramagnetic susceptibility data on the specimen containing 3% Gd indicate that the distribution of the Gd³⁺ ions is uniform even on small scales of the order of the mean distance between impurity atoms. As regards the presence in the specimen of extraneous modifications of the compound of lanthanum with indium, it follows from the results of metallographic and x-ray studies that such modifications amount to no more than 4%. The presence of 4% or less of the normal phase in the investigated specimens cannot appreciably distort the observed temperature dependence of ΔH in the superconductor since, as the results of special measurements show, the parasitic signal would not be strong enough to compete with the intense signal from the principal superconducting phase. It should also be borne in mind that the intensity of the EPR signal decreases very little (by 10–20%) at the superconducting transition point.

Thus, our experimental data permit us to say with some confidence that magnetic order and superconductivity coexist in a single phase at temperatures below 3 K in the specimens having gadolinium concentrations of 0.25 and 0.75 at.%. Since it is reasonable to assume that the temperature at which the line begins to broaden is proportional to the true magnetic ordering point, we can assert that the conditions for the appearance of magnetic order at least do not become more stringent at the transition to the superconducting phase; this becomes evident on comparing the data presented in Figs. 4 and 6. This is not even surprising, since theoretical calculations^{25,26} show that on transition to the superconducting state, the indirect exchange interactions responsible for the magnetic order do not undergo any substantial changes at distances shorter than the coherence length, which, in the investigated specimens, exceeds the average distance between localized spins.

Thus, the use of the EPR method in the present work has made it possible to detect the emergence of magnetic correlations between spins in the superconducting phase itself; the results provide grounds for concluding that in La₃In containing gadolinium impurities, a magnetically ordered phase develops in the superconducting state in practically the same way as it does in the normal state.

The authors thank T. A. Khlebnikova for assistance with the EPR measurements, and Dr. G. Wolf for performing the heat capacity measurements and discussing the results of the work.

¹Institute of Physical Problems, Academy of Sciences of the USSR, Moscow.

²Kazan State University.

³Preliminary results were published in Ref. 10.

⁴These measurements were made at the International Laboratory of Strong Magnetic Fields and Low Temperature in Wrocław, Poland.

⁵We note that for this potential, the form of the function $\chi(T)$

depends strongly on the value of k_f and when $k_f \gg a^{-1}$ (a is the lattice constant) it is not permissible to replace the summation over the lattice in formula (1) by an integration over a continuum, as was done in Ref. 14.

⁶It proved to be impossible to determine the magnitude and sign of the exchange integral from the shift of the g factor from its value for the free ion ($g_0 = 1.99$) because of the large width of the resonance line.

¹T. S. Al'tshuler, I. A. Garifullin, and E. G. Kharakhash'yan, *Fiz. Tverd. Tela* **14**, 263 (1972) [*Sov. Phys. Solid State* **14**, 213 (1972)].

²C. Rettori, D. Davidov, P. Chaikin, and R. Orbach, *Phys. Rev. Lett.* **30**, 437 (1973).

³U. Engel, K. Baberschke, G. Koopmann, S. Hufner, and M. Wilhelm, *Solid State Commun.* **12**, 977 (1973).

⁴K. Baberschke, U. Engel, and S. Hufner, *Solid State Commun.* **15**, 1101 (1974).

⁵D. Davidov, C. Rettori, and H. M. Kim, *Phys. Rev.* **B9**, 147 (1974).

⁶N. E. Alekseevskii, I. A. Garifullin, B. I. Kochelaev, and E. G. Kharakhash'yan, *Pis'ma Zh. Eksp. Teor. Fiz.* **18**, 323 (1973) [*JETP Lett.* **18**, 189 (1973)].

⁷V. I. Kochelaev, E. G. Kharakhash'yan, I. A. Garifullin, and N. E. Alekseevsky, *Proceedings of the Eighteenth Congress AMPERE*, Nottingham, 1974, p. 23.

⁸M. B. Maple, *Appl. Phys.* **9**, 179 (1976).

⁹S. Roth, *Appl. Phys.* **15**, 1 (1978).

¹⁰N. E. Alekseevskii, I. A. Garifullin, B. I. Kochelaev, and E. G. Kharakhash'yan, *Pis'ma Zh. Eksp. Teor. Fiz.* **24**, 540 (1976) [*JETP Lett.* **24**, 498 (1976)].

¹¹J. E. Crow and R. D. Parks, *Phys. Rev. Lett.* **21**, 378 (1966).

¹²J. E. Crow, R. P. Guertin, and R. D. Parks, *Phys. Rev. Lett.* **19**, 77 (1967).

¹³R. M. Seanlan and J. D. Livingston, *J. Appl. Phys.* **43**, 639 (1972).

¹⁴A. I. Lapkin and D. E. Khmel'nitskii, *Zh. Eksp. Teor. Fiz.* **58**, 1789 (1970) [*Sov. Phys. JETP* **31**, 958 (1970)].

¹⁵F. Heiniger, E. Bucher, J. P. Maita, and P. Descouts, *Phys. Rev.* **B8**, 3194 (1973).

¹⁶P. Descouts, B. Perrin, A. Dupanloup, and A. Treyvand, *J. Phys. Chem. Solids* **39**, 161 (1978).

¹⁷Yu. M. Ivanchenko, A. I. Kozinskaya, and A. A. Lisyanskiĭ, *Fiz. Tverd. Tela* **19**, 1298 (1977) [*Sov. Phys. Solid State* **19**, 755 (1977)].

¹⁸K. D. Siegel, G. Wolf, K. Bohmhammel, and H. G. Schmidt, *Exp. Technik der Physik* **25**, 229 (1977).

¹⁹L. P. Gor'kov, *Zh. Eksp. Teor. Fiz.* **37**, 1407 (1959) [*Sov. Phys. JETP* **10**, 998 (1960)].

²⁰T. S. Al'tshuler, I. A. Garifullin, E. G. Kharakhash'yan, and L. F. Shatrukov, *Fiz. Tverd. Tela* **14**, 2555 (1972) [*Sov. Phys. Solid State* **14**, 2213 (1973)].

²¹Hiroshi Hasegawa, *Progr. Theor. Phys.* **21**, 483 (1959).

²²A. A. Abrikosov and L. P. Gor'kov, *Zh. Eksp. Teor. Fiz.* **39**, 1781 (1960) [*Sov. Phys. JETP* **12**, 1243 (1961)].

²³R. P. Guertin, J. E. Crow, and R. D. Parks, *Phys. Rev. Lett.* **16**, 1095 (1966).

²⁴K. H. Bennemann, J. W. Garland, and F. M. Mueller, *Phys. Rev. Lett.* **23**, 1503 (1969).

²⁵N. E. Alekseevskii, I. A. Garifullin, B. I. Kochelaev, and E. G. Kharakhash'yan, *Zh. Eksp. Teor. Fiz.* **72**, 1523 (1977) [*Sov. Phys. JETP* **45**, 799 (1977)].

²⁶B. I. Kochelaev, L. P. Tagirov, and M. G. Khusainov, *Zh. Eksp. Teor. Fiz.* **76**, 578 (1979) [*Sov. Phys. JETP* **49**, 291 (1979)].

Translated by E. Brunner