Ordering of magnetic impurity in a superconductor

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Electron paramagnetic resonance is used to establish the existence of magnetic order of localized states of gadolinium in superconducting $La_{3-x}Gd_xIn$.

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The problem of magnetic ordering of impurities in a superconducting compound has been attracting attention for a long time. In experimental investigations of this equation, electron paramagnetic resonance (EPR) may turn out to be effective, since it makes it possible to study the interactions between the paramagnetic impurities directly in the superconducting phase. The EPR method, in particular, makes it possible to follow continuously the transition from the paramagnetic phase into the region of magnetic order. In this paper we report preliminary results of an investigation of EPR in the compound $La_{3-x}Gd_xIn$ in the normal and in the superconducting states. The choice of $La_{3-x}Gd_xIn$ is governed by the fact that earlier investigations[1,2] give grounds for assuming the possible existence of superconductivity and magnetic order in this system.

The samples for the investigations were prepared in an induction furnace in an atmosphere of pure helium and then annealed at 600 °C for 24 hours. An x-ray analysis has shown the presence of only one phase with Cu_3Au-type lattice with a constant $a = 5.07$ Å, corresponding to the compound La_3In.[3] Measurements of the temperature of the superconducting transition T_c gave a transition width 0.4 °K, thus indicating that the samples were of good quality. The dependence of the superconducting transition temperature T_c on the gadolinium impurity concentration c turned out to be linear with $\partial T_c/\partial_c \approx 0.4 K$/at.%, in good agreement with the result of[1].

The EPR measurements were made at a frequency 9300 MHz in the temperature range 1.6—30 °K. The Gd concentration in the samples ranged from 0.125 to 1.25 at.%. The temperature dependence of the line width for the investigated samples is shown in Fig. 1. It is described by the expression $\Delta H = a + bT$. The part of the line width not connected with the field of the vortices is indicated in the superconducting region.[3)]

Normal state. The character of the presented experimental results indicate that the "electron bottleneck" conditions are realized in the normal state. The temperature slope of the EPR line width is determined by the expression[5]

$$b = \frac{\pi k_B}{g \mu_B} N^2 J^2 \left[\frac{\delta_{SO}}{\delta_{ei} + \delta_{SO}} \right], \tag{1}$$

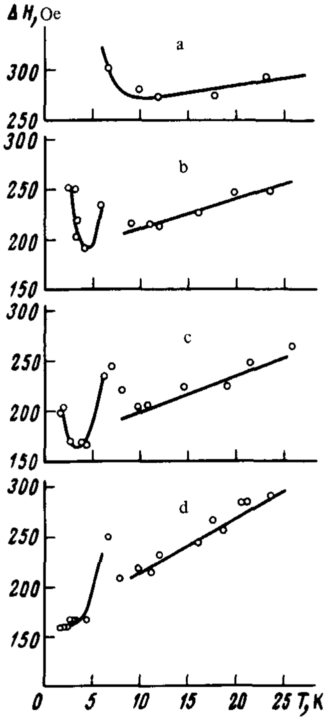

FIG. 1. Temperature dependence of the width of the EPR line for the investigated La$_{3-x}$Gd$_x$In samples: (a) $x=0.05$, (b) $x=0.03$, (c) $x=0.01$, (d) $x=0.005$. $x=0.04$ corresponds to 1 at.%. The superconducting transition temperature ranges from 9.0 to 8.6 °K with increasing gadolinium concentration.

where k_B is Boltzmann's constant, μ_B is the Bohr magneton, g is the spectroscopic splitting factor, N is the density of states of the conduction electrons with given spin orientation on the Fermi surface, J is the exchange-interaction integral of the conduction electrons and the localized moments of Gd, and δ_{ei} and δ_{SO} are the rates of exchange and spin-orbit scattering of the conduction electrons. Using the formula of Abrikosov and Gor'kov for the dependence of T_c on the concentration of an impurity with spin S[6]

$$\Delta T_c = -\frac{\pi^2}{8k_B} cNJ^2S(S+1)$$

and expression (1), we obtain $\delta_{ei}=6.7\times10^{11}c\text{-sec}^{-1}$, $\delta_{SO}=2.8\times10^{11}\ \text{sec}^{-1}$, $N=0.26\ \text{eV}^{-1}\text{atom}^{-1}\text{spin}^{-1}$, and $J=0.07$ eV.

For a sample with the largest Gd concentration ($x=0.05$) in the normal region, a broadening and a shift of the resonance line is observed at 9 °K in the direction of weaker magnetic fields. This is a typical manifestation of the magnetic ordering the EPR spectra (see, e.g., [7]). The broadening of the EPR line following magnetic ordering is due to the abrupt increase of the susceptibility, which leads to an increase of the relaxation of homogeneous precession of the localized moments.

Superconducting state. At gadolinium concentrations $x<0.05$, when the EPR measurements can be carried out in the superconducting state, one observes the usual increase of the line width near T_c, due to the coherence effects. At temperatures ~3 °K, for samples with Gd concentrations $x = 0.03$ and 0.01, a line broadening takes place, analogous to the broadening for the sample with $x = 0.05$ in the normal state. This offers evidence of magnetic ordering in the superconducting phase.

Our data do not make it possible at present to establish the type of magnetic order in the investigated system, or to determine, in particular, whether it is homogeneous in space or has a cluster character, similar to the "mictomagnetic" or "spin glass" (see, e.g., [8]). A continuation of these investigations, namely a comparison of the ordering curves in the normal and superconducting states (measurement of the EPR at different frequencies) and introduction of an additional spin-orbit scatterer will cast light on the question of the question of the nature of ordering in the superconducting phase.

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(3) The procedure of determining ΔH from the experimental spectra in the superconducting phase is described in[4].

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[1] J. H. Crow and R. D. Parks, Phys. Lett. **21**, 378 (1966).
[2] J. H. Crow, R. P. Guertin, and R. D. Parks, Phys. Rev. Lett. **19**, 77 (1967).
[3] B. T. Mattias, V. B. Compton, and E. Corenzwit, J. Phys. Chem. Solids **19**, 130 (1961).
[4] H. Hasegawa, Prog. Theor. Phys. **27**, 483 (1959).
[5] B. I. Kochelaev, E. G. Kharakhash'yan, I. A. Garifullin, and N. E. Alekseevsky, 18th Ampere Congress, Nottingham, 23, 1974.
[6] A. A. Abrikosov and L. P. Gor'kov, Zh. Eksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. JETP **12**, 1243 (1961)].
[7] R. H. Taylor and B. R. Coles, Metal Phys. **5**, 121 (1975).
[8] J. A. Mydosh, Twentieth Annual Conference on Magnetism and Magnetic Materials, San Francisco, 1974.