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CONDENSED MATTER

Magnetic Resonance Spectrum of a Two-Phase State in Single Crystals of $La_{0.7}Pb_{0.3}MnO_3$ Lanthanum Manganite

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Abstract—Two absorption lines are observed over a wide temperature range below T_c in the magnetic resonance spectrum of an $La_{0.7}Pb_{0.3}MnO_3$ single crystal. These lines correspond to two magnetic phases in the sample. The frequency–field dependence of spectra obtained in the range of microwave radiation frequencies 10–77 GHz allows these phases to be interpreted as ferromagnetic and paramagnetic phases. The phase volume ratio depends on the temperature and the magnitude of the external magnetic field. Features in the temperature behavior of parameters of the magnetic absorption line are observed in the region of the highest magnetic resistance of the sample. The results are interpreted within the mechanism of electronic phase separation.

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At present, the mechanism of electronic phase separation is one of the most widely discussed mechanisms of colossal magnetic resistance in ferromagnetic manganites with the perovskite structure [1]. Direct evidence for the coexistence in the material bulk of a ferromagnetic metallic phase and a phase with localized charge carriers was obtained in experiments on studying NMR [2], optical properties [3], electron diffraction [4], and neutron scattering [5]. It is evident that such a mixed two-phase state must be manifested in an investigation by the magnetic resonance method, which is rather sensitive to the magnetic inhomogeneity of materials. Moreover, because the two-phase state in electronic separation is controlled by the external magnetic field, magnetic resonance spectra recorded at different frequencies of microwave radiation can differ significantly. The few previous investigations of magnetic resonance in lanthanum manganite materials were ambiguous, and the problem was stated differently in these works. At the same time, the results of almost all works gave evidence of the magnetic inhomogeneity of the materials under study, although these materials were homogeneous in the crystallographic respect [6, 7].

In this work, we report the results of investigating the magnetic resonance spectra of $La_{0.7}Pb_{0.3}MnO_3$ single crystals, which exhibit the effect of colossal magnetic resistance. Measurements were carried out on spectrometers with (1) the working frequency $\nu = 10$ GHz and a constant magnetic field and (2) a tunable frequency in the range $\nu = 37$–77 GHz and a pulsed magnetic field. Single crystals of $La_{0.7}Pb_{0.3}MnO_3$ were grown by spontaneous crystallization from a molten mixture [8]. The samples under study were shaped like thin plates ($4 \times 2 \times 0.1$ mm) whose plane coincided with one of the main crystal planes. The magnetic field was applied along the plane of the sample.

The main feature of the magnetic resonance spectra in the crystals under study is the occurrence of two well-resolved lines of magnetic absorption in a wide temperature range. The temperature behavior of the parameters of absorption lines for the frequency $\nu = 10$ GHz is shown in Fig. 1. We believe that the line designated in Fig. 1 as *F* corresponds to a resonance from the crystal domains that occur in the ferromagnetic state. Line *F* appears only below the Curie point $T_c \approx 360$ K; and its intensity increases with decreasing temperature, which may be associated with an increase in crystal magnetization M_0 and also with an increase in the volume of the ferromagnetic phase in the sample. A decrease in the resonance field H_r^F with decreasing temperature is associated with anisotropic interactions, that is, with a rise in magnetic crystallographic anisotropy and with sample shape anisotropy. Additional orientational investigations of the magnetic resonance in samples of spherical shape showed that the effective field of magnetic crystallographic anisotropy H_A does not exceed 100 Oe. Variations of H_r^F are mainly associated with a rise in degaussing fields, which are proportional to the magnetic moment. The following equation may be written for the geometry in which our experiment was carried out ($H_{0\|}$ is parallel to the sample plane):

$$\omega/\gamma = (H_0 + 4\pi M_{\text{eff}})^{1/2}, \quad (1)$$

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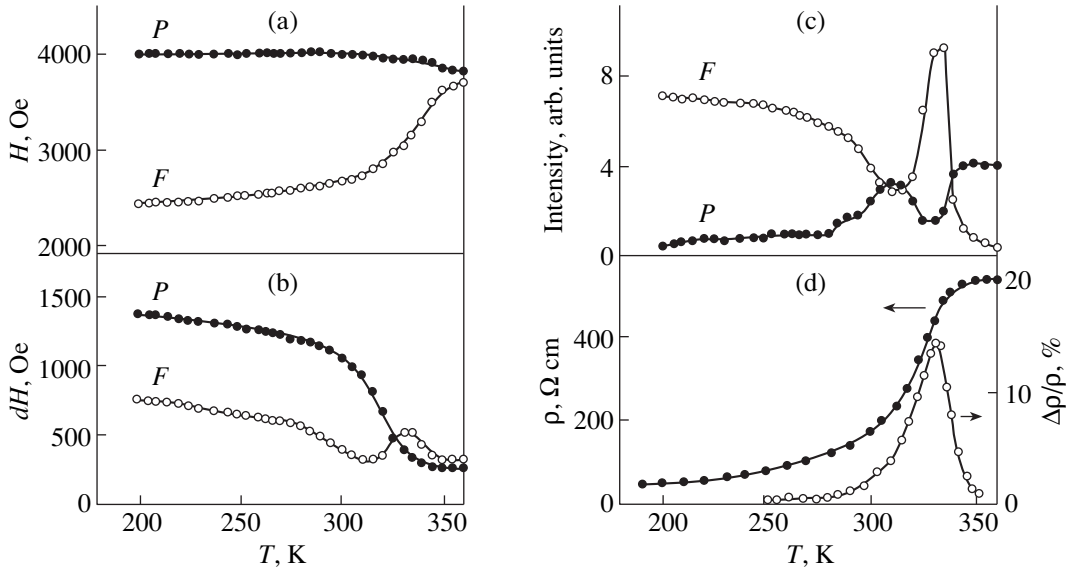

Fig. 1. Temperature dependence of the parameters of lines *F* and *P* in the magnetic resonance spectrum of an $La_{0.7}Pb_{0.3}MnO_3$ single crystal (ν = 10 GHz): (a) resonance field, (b) line width, and (c) line intensity; (d) temperature dependence of the resistivity ρ_0 and the magnetic resistance $\Delta\rho/\rho_0$ in the field H = 7 kOe.

where $\omega = 2\pi\nu$ is the angular frequency, $\gamma = (g\mu_B/\hbar)$ is the gyromagnetic ratio, H_0 is the resonance field, and M_{eff} is the effective magnetization. In the general case, M_{eff} will differ from M_0 because of the effect of crystallographic anisotropy, which is neglected in (1), and also because of possible magnetic inhomogeneity of the crystal (which, as will be shown below, does occur). At the same time, the temperature behavior of the quantity $4\pi M_{\text{eff}}(T)$ determined from (1) qualitatively repeats the behavior of the quantity $4\pi M_0(T)$ obtained from static measurements.

Line *P*, which is observed in the paramagnetic state of the sample ($T > T_c$), virtually retains its position as the temperature decreases below T_c, and its resonance field H_r^P does not depend on the sample shape and the orientation of H_0 in the crystal. The insignificant increase in H_r^P in the temperature range 340–350 K may be associated with the change in the *g*-factor due to local distortions of the crystal lattice. Such distortions arising from strong electron–phonon coupling were revealed for a number of compositions of impurity manganese perovskites in the temperature range corresponding to the transition to the ferromagnetic state [9]. The decrease in the intensity of line *P* with decreasing temperature indicates that the fraction of the paramagnetic phase in the sample decreases.

A significant broadening of the ferromagnetic resonance line dH_r^F with decreasing temperature has also been observed previously by various authors in studying crystalline and film manganite samples. However, a thorough analysis of the mechanisms leading to an increase in dH_r^F has not been presented yet. From our point of view, the electrical conductivity of the samples is the main factor that affects dH_r^F. Actually, the resistivity ρ of the crystals rapidly drops below T_c (Fig. 1). This means an increase in either the mobility or the concentration of charge carriers, which, in its turn, exerts a significant effect on dynamic magnetic phenomena. The contribution of charge carriers to dissipation processes can have several mechanisms as its basis [10]. An inhomogeneous two-phase magnetic state must also exert a significant effect on dH_r^F. This state will lead to nonuniform magnetization of ferromagnetic domains and, consequently, to a broadening of the ferromagnetic resonance line.

The features in the behavior of magnetic absorption line intensities and widths at a temperature $T \sim 325$ K engage the attention. It is interesting that the maximum in the temperature dependence of $\Delta\rho/\rho$ corresponds to just the same temperature (Fig. 1).

Undoubtedly, the behavior of the frequency–field dependence for the absorption lines (Fig. 2) is an important argument in favor of the statement that the observed lines *P* and *F* correspond to resonance absorption from paramagnetic and ferromagnetic domains, respectively. An extrapolation of the experimental points in the frequency–field dependence for line *P* indicates that the dependence is linear and passes through the origin of the coordinates. This pattern is observed for any orientation of the external magnetic field H_0. This fact confirms that line *P* is associated with the domains in the crystal that occur in the paramagnetic state even below T_c. The behavior of the fre-

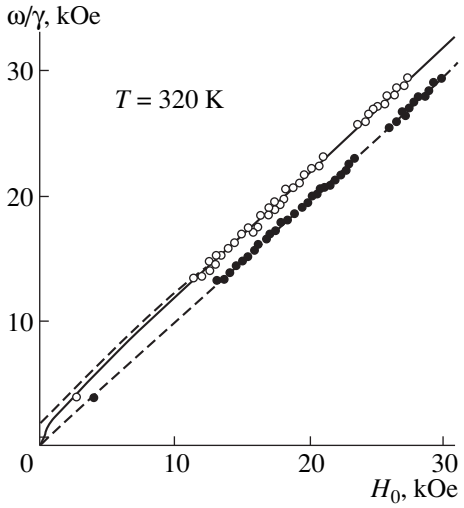

Fig. 2. Frequency–field dependence of lines *F* and *P* in the magnetic resonance spectrum of an $La_{0.7}Pb_{0.3}MnO_3$ crystal, $T = 320$ K. Solid line represents dependence (1) (see text), and dashed lines are linear extrapolations of experimental points.

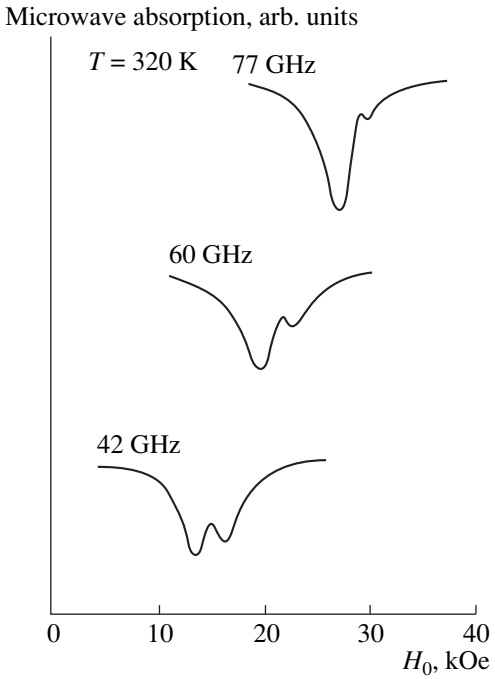

Fig. 3. Magnetic resonance absorption spectra of an $La_{0.7}Pb_{0.3}MnO_3$ crystal recorded at various microwave radiation frequencies ν (42, 60, and 77 GHz); $T = 320$ K.

quency–field dependence for line *F* is characteristic of uniform magnetic oscillations in a ferromagnetic material. The shift of the dependence with respect to the origin is determined in our case mainly by the anisotropy of the sample shape (see (1)).

The dependence of the intensities of lines *P* and *F* on the microwave radiation frequency at which the magnetic resonance spectrum has been recorded is another important experimental result in addition to the demonstration of the two-phase paramagnetic–ferromagnetic state (Fig. 3). If the frequency ν and, hence, the magnitude of the external magnetic field H_0 were increased at a fixed temperature, the intensity of line *F* increased and that of line *P* decreased. This fact can be interpreted as control of the ratio between the phase volumes by the external magnetic field. An increase in H_0 gives rise to an increase in the volume of the ferromagnetic phase and to a decrease in the volume of the paramagnetic phase in the sample. It should be noted that some other factors, for example, a change in the value of magnetization M_0, can also contribute to the intensity of the ferromagnetic resonance absorption line. However, the frequency–field dependence for line *F* is described well by equation (1) with a constant value $4\pi M_{\text{eff}} \approx 2$ kOe (see Fig. 2). Hence, the ferromagnetic domains in the crystal in magnetic fields used for measurements were magnetized to saturation.

As noted above, the mechanism of electronic phase separation is considered one of the possible mechanisms responsible for colossal magnetic resistance in lanthanum manganites. In this case, domains with various concentrations of carriers form in a crystal homogeneous in the crystallographic (chemical) sense. These domains differ in their conductivity and magnetic state. This heterophase state of the crystal corresponds to the state with minimal energy, that is, to the ground state. The inhomogeneous two-phase state must be very sensitive to the action of external parameters, for example, temperature, and the external magnetic field, which provides an explanation for the high value of magnetic resistance.

The occurrence of features in the width and intensity of magnetic resonance lines in the region of the highest magnetic resistance confirms that the two-phase magnetic state observed in this work is connected with anomalous magnetoresistive properties in $La_{0.7}Pb_{0.3}MnO_3$ single crystals (Fig. 1).

The situation when magnetic impurity phase separation is accomplished because the impurity is nonuniformly distributed in the process of synthesis of the crystals cannot be excluded. However, strong effects of the external magnetic field can hardly be expected in this case.

Thus, a two-phase magnetic (paramagnetic–ferromagnetic) state controlled by the magnitude of the external magnetic field was observed in $La_{0.7}Pb_{0.3}MnO_3$ single crystals by the magnetic resonance method.

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