Resonant microwave conductivity response to ac current in $La_{0.7}Pb_{0.3}MnO_3$ crystals

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An experimental study of the effect of low-frequency transport current on the microwave conductivity of single-crystal $La_{0.7}Pb_{0.3}MnO_3$ is reported. In the absence of an external magnetic field, the microwave conductivity response to a current follows a relaxation behavior. In a nonzero external magnetic field, the response spectrum exhibits a peak of resonant amplitude enhancement. The resonant response has a nonlinear nature. The temperature and field dependences of the main parameters of the microwave response correlate directly with the behavior of the magnetoresistance. The results obtained are analyzed within the oscillator approximation. Electronic phase separation is proposed as a possible mechanism for the current action. [S1063-7834(99)01612-3]

Perovskite-structure manganites with a common formula $R_{1-x}A_xMnO_3$ (where R stands for rare-earth ions, and A stands for Sr, Ba, Ca, and Pb) have been recently attracting attention due to their unusual magnetic and electronic properties.[1] Most of this interest centers on the giant magnetoresistance (GMR) observed in some compositions.[2] At the same time there is still no full understanding of the mechanisms responsible for the GMR and other magnetoelectric effects. A substantial contribution to the solution of this problem could come from invoking nontraditional experimental methods, for instance, those aimed at studying the response of a system to combined action of various factors. Besides, one could expect in this case new manifestations of the GMR, as well as new effects having application potential.

In this connection, the microwave conductivity response to an ac current in single-crystal $La_{0.7}Pb_{0.3}MnO_3$ exhibiting GMR, which was detected by us,[3] turned out to be very interesting. The nature of the response depended on the amplitude of the ac voltage across the sample and its frequency, as well as on the external magnetic field. This observation is a one more demonstration of an intimate relation between the magnetic and electrical properties of perovskite-type manganese oxides. This communication reports the results of a detailed investigation of this phenomenon.

1. EXPERIMENTAL

The studies were carried out on $La_{0.7}Pb_{0.3}MnO_3$ single crystals grown by spontaneous crystallization from a melt solution.[3] The sample representing a polished plate measuring $4\times2\times0.1$ mm^3 was placed at the antinode of a microwave electric field in a rectangular cavity ($\nu=10$ GHz). The cavity was connected in a reflection-type magnetic-resonance spectrometer arrangement. The electric current was fed into the sample through spring-loaded needle contacts. One measured the microwave power P_c reflected from the cavity with the sample as a function of temperature, external magnetic field, and the frequency and amplitude of the ac voltage across the sample.

It is well known[4] that if the sample dimensions are much smaller than the wavelength of the electromagnetic radiation, and the skin-layer thickness is larger than or comparable to the smallest sample dimension (in our case this condition was met), the variation of P_c can be related in a single-valued way to that of the complex dielectric permittivity

$$\varepsilon=\varepsilon'-i\varepsilon''=\varepsilon'-i\frac{\sigma_{MW}}{\omega}, \quad (1)$$

where σ_{MW} is the conductivity in the microwave range and ω is the wavelength of the electromagnetic radiation. For conducting media one usually has $\varepsilon'\ll\varepsilon''$. Indeed, in our experiments we did not find any variation of the cavity resonance frequency which could be related to that of ε'. The microwave response signal was generated by variations of the cavity Q-factor. The relative change in the Q-factor caused by a change in the conductivity $\Delta\sigma_{MW}$ can be written[5]

$$\frac{\Delta Q}{Q_0}=\frac{Q_0}{\omega_0 W_0}\Delta\sigma_{MW}\int_{V_S}E^2dV. \quad (2)$$

Equation (2) was derived by the perturbative technique, $Q_0=\omega_0W_0/(P_s+P_r)$ is the intrinsic Q-factor of the cavity complete with the sample, P_r and P_s are, respectively, the microwave power losses in the cavity walls and the sample, W_0 is the power stored in the cavity, and ω_0 is the cavity resonance frequency. The output signal of the spectrometer linear microwave detector is related to the variation of the microwave conductivity $\Delta\sigma_{MW}$ through

$$\frac{\Delta U}{U_0}=\frac{1}{2}\frac{\Delta Q}{Q_0}, \quad (3)$$

where U_0 is the microwave-detector output voltage, which is proportional to the power incident on the cavity P_{in}, and

ΔU is the change of the microwave-detector output voltage caused by a change in σ_{MW}. Finally we have

$$\Delta U = c\Delta\sigma_{MW}, \tag{4}$$

where c is a quantity remaining constant during the measurement.

Determination of the absolute value of σ_{MW} meets with certain difficulties, but we were interested in the variation of the conductivity $\Delta\sigma_{MW}$ caused by an external action. $\Delta\sigma_{MW}$ was measured in arbitrary units. For definiteness, we shall call in what follows the variation of the microwave conductivity generated by the action of a current the microwave response of a sample.

The dc resistivity ρ and the magnetoresistance $[\rho(0) - \rho(H)]/\rho_0 = \Delta\rho/\rho_0$ were measured by the standard four-probe technique.

2. EXPERIMENTAL RESULTS

Our study of the effect of transport current on the conductivity of $La_{0.7}Pb_{0.3}MnO_3$ single crystals in the microwave range revealed the following. If one applied across a sample an ac voltage $U_\sim$ of frequency f, the microwave response signal corresponding to $\Delta\sigma_{MW}$ could be represented as a sum of harmonic components of the modulating voltage frequency

$$\Delta\sigma_{MW}(t) = \sum_{i=1}^{n} A_i \cos(ift), \tag{5}$$

where i is the harmonic number, and A_i is the amplitude of the ith harmonic component in the response signal.

In the absence of an external magnetic field ($H=0$), the microwave response signal contains only even harmonics, with the main contribution being due to the component of the $2f$ frequency. This is a direct consequence of the response being independent of the polarity of the voltage applied to the sample. The temperature dependence of $\Delta\sigma_{MW}$ was presented in Ref. 3. We may recall that it coincided completely with the behavior of the sample magnetoresistance. As for the dependence of the microwave response signal on the frequency f of the applied voltage U, it is shown in Fig. 1. The data are given for $T=300$ K. We see that σ_{MW} is most sensitive to low-frequency currents. The amplitude of $\Delta\sigma_{MW}$ falls off rapidly with increasing frequency.

When an external magnetic field is applied, the response signal amplitude decreases slightly throughout the frequency range covered while remaining a smooth function of f. At the same time at a certain frequency f_0 there appears a peak of the resonantly enhanced microwave-response amplitude. Note that it is the first harmonic [see Eq. (5)] of the signal, which was absent in the $H=0$ case, that contributes to the response enhancement. As H increases, the amplitude of the resonance peak grows to exceed considerably the $H=0$ response. Indeed, at $T=300$ K and a field $H=7$ kOe the signal increases by nearly 10 times. Besides the direct resonant change $\Delta\sigma_{MW}$ at frequency f_0, the spectrum contains also harmonics of higher frequencies multiples of the current frequency. Namely, one detected sample response peaks at frequencies $f_0/3$ and $f_0/5$, with higher-order multiples also observed sometimes.

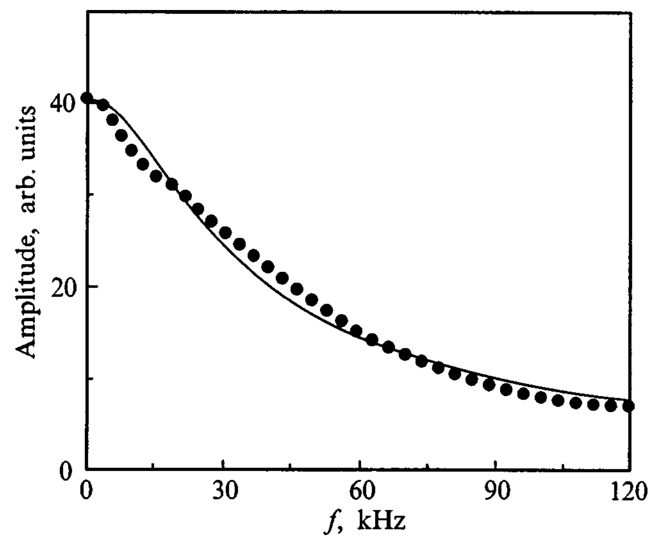

FIG. 1. Amplitude of the microwave-conductivity response signal $\Delta\sigma_{MW}$ vs frequency f of the ac voltage U acting on a sample. External magnetic field $H=0$, $U_\sim=500$ mV, $T=300$ K. Solid line: approximation (see text for explanation).

We studied the behavior of the resonant microwave-response enhancement in a field $H=7$ kOe at different temperatures (Fig. 2). In these measurements, the voltage across

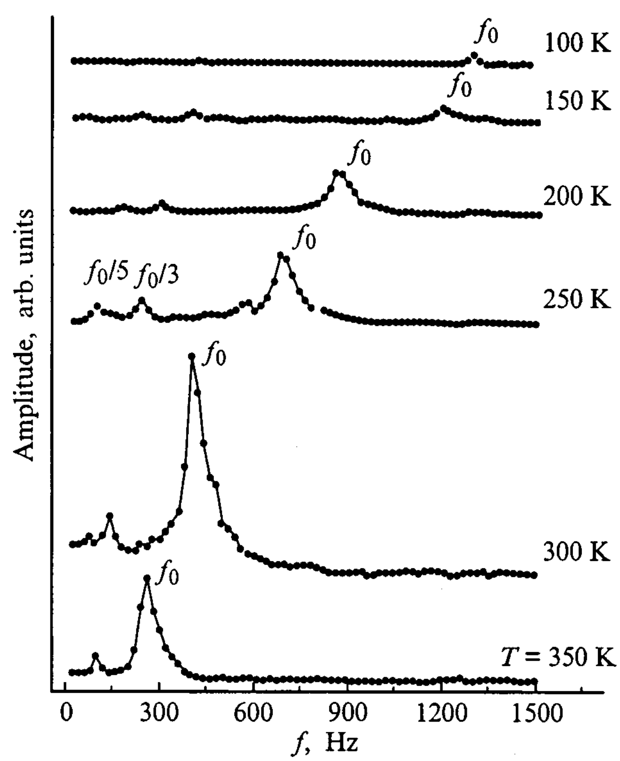

FIG. 2. Amplitude of the microwave-conductivity response signal $\Delta\sigma_{MW}$ vs the frequency f of the voltage $U_\sim$ applied across the sample placed in a magnetic field $H=7$ kOe. Different curves are plots obtained at different temperatures for $U_\sim=500$ mV. f_0 is the main-peak frequency; also shown are peaks of the sample response to frequency multiples of f_0.

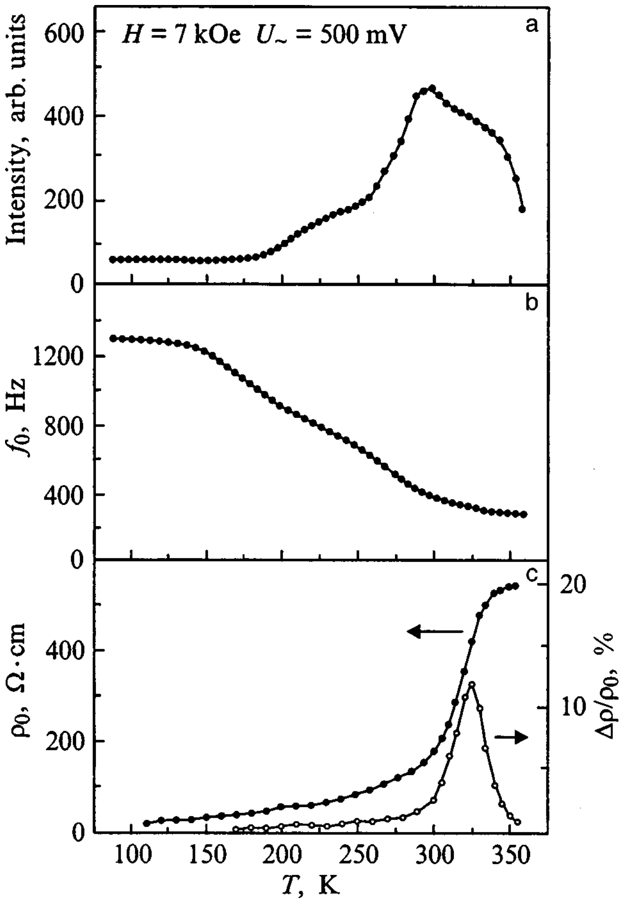

FIG. 3. Temperature dependences of the intensity (a) and frequency (b) of the peak in the microwave-conductivity resonance response $\Delta\sigma_{MW}$ of a sample to an ac voltage; $U_{\sim}=500$ mV, $H=7$ kOe; (c): temperature dependence of the resistivity ρ_0 and magnetoresistance $\Delta\rho/\rho_0$ in a field $H=7$ kOe.

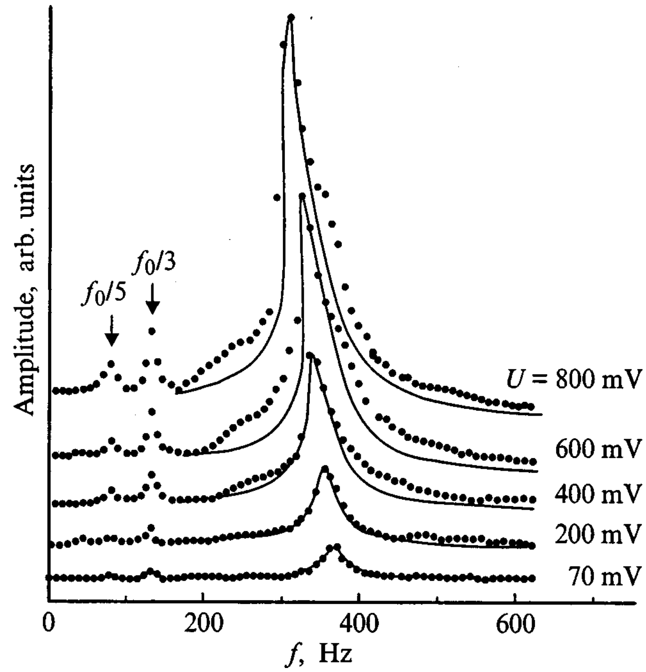

FIG. 4. Microwave-conductivity resonance response $\Delta\sigma_{MW}$ obtained for different values of $U_{\sim}$ in an external magnetic field $H=7$ kOe at $T=300$ K. Solid lines: calculations made in the nonlinear-oscillator approximation (see text for explanations).

the sample was maintained constant ($U_{\sim}=500$ mV). The peak in the dependence of $\Delta\sigma_{MW}$ on the frequency f of the voltage acting on the sample becomes noticeable at $T\sim100$ K. As the temperature increases, the peak grows in intensity and shifts toward lower frequencies. The peak of the resonant $\Delta\sigma_{MW}$ amplitude enhancement reaches the maximum intensity at about $T\sim300-320$ K. The peak amplitude decreases rapidly with a further increase of temperature. The variation of the resonant response frequency f_0 is the strongest within the temperature region from 150 to 300 K. Figure 3a and 3b illustrates graphically the phenomenon. Note the coincidence of the main features in the above-mentioned relations with the behavior of the magnetoresistance $\Delta\rho/\rho_0$ in the crystals studied (Fig. 3c). The peak of the resonance response reaches a maximum at $T\sim300$ K, where $\Delta\rho/\rho_0$ starts to grow rapidly. The drop in the response intensity is accompanied by a decrease of $\Delta\rho/\rho_0$. None of these two effects is observed for $T>T_c\sim360$ K.

Figure 4 presents a part of the family of resonant microwave response curves obtained by varying the ac voltage $U_{\sim}$ across the sample. The temperature $T=300$ K and the magnetic field $H=7$ kOe were fixed. One readily sees that as $U_{\sim}$ increases, the resonance line increases in intensity too, and becomes distorted. The maximum of the curve shifts toward lower frequencies. The peaks at the $f_0/3$ and $f_0/5$ frequencies do not change position in the spectrum. This behavior is typical of forced oscillations in nonlinear systems under harmonic excitation. It can be added that at large $U_{\sim}$ amplitudes the resonance curve exhibits a slight hysteresis with increasing and decreasing frequency f. This deviation of the resonance curve from single-valuedness is also a consequence of the nonlinearity of the system under study.

Interestingly, if one fixes $U_{\sim}$ at a given temperature but varies the external magnetic field, a similar family of resonance curves is obtained (Fig. 5). This permits a conclusion that the amplitude of the exciting force depends on a combination of the quantities H and $U_{\sim}$.

Note that different samples could differ slightly in the magnitude of f_0, the width and shape of the resonant response peaks at a fixed temperature. At the same time the pattern of the above effects did not change in any way. We associate the observed differences with the quality of the single crystals used, including nonuniform impurity distribution over the sample. This conclusion is corroborated indirectly by studies of the magnetic resonance in the crystals.

3. DISCUSSION

The influence itself of both dc and ac transport current on the electrical properties of $La_{0.7}Pb_{0.3}MnO_3$ single crystals turned out to be unexpected. The observed effect is apparently one more manifestation of the unusual electrical and magnetic properties of perovskite-like manganese oxides, so that the mechanisms of the current action and of the GMR in the crystals under study have the same nature. This is supported, first, by all the main features found in the study of the

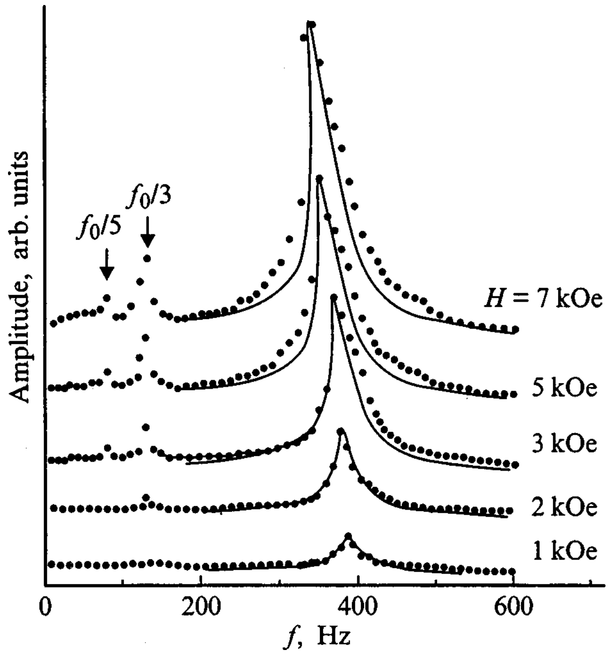

FIG. 5. Microwave-conductivity resonance response $\Delta\sigma_{MW}$ obtained for different strengths of external magnetic field, $U_{\sim} = 500$ mV, and $T = 300$ K. Solid lines: calculations made in the nonlinear-oscillator approximation (see text for explanations).

temperature dependences being correlated, and, second, by the behavior of the above effects in an external magnetic field.

In order to describe in detail the response of a system to a change in an external parameter, one has to know its internal structure, i.e., its inherent interactions and the mechanisms by which the current and magnetic field act on the state of the substance. Unfortunately, the main question of the mechanisms of the current action on microwave conductivity, as of the nature of the other magnetoelectric phenomena in manganites, remains unanswered.

On the other hand, the internal structure of a system can be judged from its response to a perturbation. Thus an analysis of the dynamic behavior of σ_{MW} as a function of the amplitude and frequency of the modulating voltage, temperature, and magnetic field may provide additional information concerning the nature of interactions in the system under study.

Consider first the $H=0$ case. The dependence of the response amplitude on the frequency of the ac voltage applied to the sample (Fig. 1) is characteristic of the case where the processes accounting for the variation of the propeties of the system occur at a finite rate. As a result, the response of the system lags behind the external perturbation (the magnetic aftereffect is a particularly revealing example of such a behavior). In the simplest case, the $\sigma_{MW}(t)$ function is determined only by one relaxation time τ_r. This can be written mathematically as follows[6]

$$\frac{\partial\sigma_{MW}}{\partial t} = -\frac{\sigma_{MW}-\sigma^{e}_{MW}}{\tau_r}, \tag{6}$$

where σ^{e}_{MW} is the equilibrium value of the microwave conductivity to which the system relaxes. We shall assume the perturbation to be small, in which case the response can, in a first approximation, be presented as[3]

$$\sigma^{e}_{MW} = \xi U^2 + \sigma_0, \tag{7}$$

where ξ is a constant, the quadratic dependence on U is a consequence of the independence of $\Delta\sigma_{MW}$ of the polarity of the applied voltage, and σ_0 is the dc component of the microwave conductivity, which is not seen in an experiment. We present the ac voltage in the form

$$U = U_0 e^{ift}. \tag{8}$$

Because of the lag, the microwave conductivity can be written

$$\sigma_{MW} = A_2 e^{i(2ft-\delta)} + \sigma_0, \tag{9}$$

where $2f$ corresponds to the response at the doubled perturbation frequency, and A_2 is the response amplitude. Substituting (7), (8), and (9) into Eq. (6) yields

$$A_2 = \frac{\xi U_0^2}{1+(2f\tau_r)^2}(\cos\delta + 2f\tau_r \sin\delta), \tag{10}$$

$$\tan(\delta) = 2f\tau_r, \tag{11}$$

The relations thus obtained fit with a good accuracy to experimental data for $\xi = 162$ ($U_{\sim} = 500$ mV) and $\tau_r = 4.4\times10^{-5}$ s.

Thus invoking only one time constant τ_r is a good enough approximation to determine the response time of a sample to an applied voltage. We have essentially employed a classical relaxation model of an overdamped oscillator, in which the friction coefficient is much larger than the characteristic oscillator natural frequency. Actually, the processes affecting the variation of the electrical properties of a material under study should most likely be characterized by a spectrum of time constants (which corresponds to an ensemble of oscillators with a certain distribution function). The approximation we have used is equivalent to introducing some average effective time constant. A more substantial conclusion could be drawn after a special investigation of the behavior of the system response to a perturbation in time.

We shall instead dwell on the microwave response of a sample to an ac current in an external magnetic field. Of particular significance here is the resonant enhancement of the response amplitude at the f_0, $f_0/3$, and $f_0/5$ frequencies, while at all other frequencies the response signal exhibits usual relaxation. Thus the dynamic behavior of the microwave conductivity of a sample in an external magnetic field is characterized by two time constants ($\tau_r \sim 10^{-3}$ s and $\tau_0 \sim 1/f_0 \sim 10^{-3}$ s) differing by two orders of magnitude. This difference permits one, in a first approximation, to consider the processes responsible for each of the times separately.

As follows from the above experimental data, we have here obviously forced oscillations in a nonlinear dynamic system. It appears only natural to choose as a first step the simplest model, i.e., a nonlinear oscillator. Within this approximation, the equation of motion for the dynamic behavior of the microwave conductivity σ_{MW} of the system under study can be written

$$\frac{d^2x}{dt^2}+2\delta\frac{dx}{dt}+f_0^2x+g(x)=P\cos(\Omega t). \quad (12)$$

Here $x=\sigma_{MW}/\sigma_n$ is a dimensionless variable, σ_n is an arbitrary normalization factor, δ is the damping parameter, f_0 is the natural frequency of a linear oscillator, P is the amplitude of the external force, Ω is its frequency, $g(x)=-\partial U(x)/\partial x$ is a function characterizing the nonlinearity, and $U(x)$ is the oscillator potential energy.

The properties of the system determining the natural oscillation frequency f_0 vary with temperature, and at a fixed temperature f_0 remains constant. This can be seen from the constancy of the peak positions at the $f_0/3$ and $f_0/5$ frequencies in the response spectrum. The variation of the oscillator frequency with oscillation amplitude is a consequence of the nonlinearity (nonisochronism). The presence in the response spectrum of odd harmonics only permits a conclusion that the $g(x)$ function must be odd. The nonlinear relations characterizing the properties of a system are usually approximated with a polynomial. Thus we can write to the seventh power of x

$$g(x)=\alpha x^3+\beta x^5+\gamma x^7. \quad (13)$$

Further analysis shows that within the model considered here δ also remains constant at a fixed temperature. The quantities $U_\sim$ and H determine the amplitude of the external force

$$P=P(U_\sim,H). \quad (14)$$

We restrict ourselves to obtaining the solution of Eq. (12) for the frequency of the external force. In the case of weak nonlinearity it can be solved, for instance, by harmonic approximation[7]

$$f^2=(f_m^2-2\delta^2)\pm\sqrt{\frac{P^2}{A^2}-4\delta^2(f_m^2-\delta^2)}, \quad (15)$$

where

$$f_m^2=f_0^2\left(1+\frac{3}{4}\alpha A^2+\frac{5}{8}\beta A^4+\frac{35}{64}\gamma A^6\right) \quad (16)$$

is the squared frequency of the same nonlinear system for $\delta=0$ and A is the amplitude. Using the skeleton curve equation (16), one can readily find the experimental values of the α, β, and γ parameters. The best fit was obtained for $\alpha=-1.25\times10^{-5}$, $\beta=2.45\times10^{-10}$, and $\gamma=-1.5\times10^{-15}$, irrespective of which external perturbation parameter, $U_\sim$ or H, was varied. Next, by fitting the solution of (15) to experimental data (Figs. 4 and 5), one obtains the form of the relation (14). Calculations show that the external force P is a linear function of both $U_\sim$ and H. Note that within the approximation employed here the theoretical resonant-response curves reproduce well enough the experimental relations.

Knowing $g(x)$, one could reconstruct the potential function of the system under study and reproduce qualitatively the complete pattern of the motion of the nonlinear oscillator. We are more interested, however, in the physical meaning of the relations obtained, because they are connected with actual interactions in the material. Unfortunately, as this was pointed out more than once, the understanding of the mechanisms involved is far from being unambiguous.

As before,[3] we espouse the viewpoint that the mechanism underlying the phenomena considered here is phase separation[2,8]. Our opinion is supported by additional magnetic-resonance studies of single-crystal $La_{0.7}Pb_{0.3}MnO_3$. Within a broad temperature range below T_c one observes two magnetic-resonance absorption lines, which correspond to two magnetic phases in the sample. Field-frequency relations permit their identification as the ferromagnetic (FM) and paramagnetic phases. Moreover, the relation between the phase volumes at a fixed temperature is governed by the external magnetic field. It is this phenomenon that underlies the mechanism accounting for the GMR.

In our case, the transport current, as well as the magnetic field, influences the volume ratio of the phases differing in the magnetic state and, accordingly, in the conductivity. This affects the microwave conductivity of a sample.[3] Incidentally, the response of a sample to the action of a current is of the opposite sign to that of the magnetic field. This can be qualitatively understood based on the following considerations. The total free energy of a two-phase system is given by the expression[9]

$$E=E_V+E_S+E_Q+E_M. \quad (17)$$

Here E_V and E_S are, respectively, the volume and surface energies of the conduction electrons, E_Q is the Coulomb interaction energy between regions with different electron concentrations, and E_M is the magnetic energy, which includes the energies of the s–d exchange, direct exchange coupling, and interaction with the external field. A variational analysis of Eq. (17) yields the equilibrium dimensions of the phase nonuniformities. External magnetic field acts on the magnetic part of the free energy, E_M, which results in an increase of the FM-phase volume of the crystal.[10] Transport current increases the kinetic energy of the conduction-band electrons (through the change of E_V and E_S), which brings about destruction of the ferromagnetism and, accordingly, a decrease of the FM-phase volume in the crystal.

The response of a system to ac current is governed by the dynamics of phase volume variation. In this case the frequency f_0 should apparently reflect the characteristic size of the phase nonuniformity. As the temperature increases, the volume of the minor phase increases and, as a result, the frequency f_0 decreases, while the resonance response grows in intensity (Fig. 3). Above T_c, the sample becomes spatially uniform.

Within the oscillator approach, a two-phase system can be described in terms of the bistable oscillator model. In this case the presence of two characteristic time scales, τ_r and τ_0, can be explained as follows. The relaxation behavior corresponds to motion in the vicinity of one of the equilibrium states (intrawell dynamics), whereas the other scale characterizes the average time required to cross the potential barrier (global dynamics).

Thus our experimental studies have revealed two characteristic scenarios of the behavior of microwave conductivity in single-crystal $La_{0.7}Pb_{0.3}MnO_3$ samples acted upon by an ac current. In the absence of an external magnetic field, the response has a relaxation pattern with a characteristic relaxation time $\tau_r \sim 10^{-5}$ s. When placed in an external magnetic field, the low-frequency response spectrum exhibits, superposed on the relaxation behavior, peaks of resonantly enhanced response amplitude. An analysis of the results made in terms of the oscillator approach yielded the parameters of the system determining the nature of possible interactions, which are responsible for the observed magnetoelectric effects.

It may be pointed out in conclusion that the temporal behavior of the conductivity of manganites subjected to a magnetic field presents a certain interest. As far as we know, no relevant studies have thus far been carried out, although this aspect of the problem is crucial from the standpoint of applications of the GMR effect.

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