SEMICONDUCTORS

Change in the Magnetocapacity in the Paramagnetic Region in a Cation-Substituted Manganese Selenide

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Abstract—The capacity and the dielectric loss tangent of a $Gd_xMn_{1-x}Se$ ($x \leq 0.2$) solid solution have been measured in the frequency range $1-300$ kHz without a magnetic field and in a magnetic field of 8 kOe in the temperature range $100-450$ K, and the magnetic moment of the solid solution has been measured in a field of 8.6 kOe. The magnetocapacity effect and the change in the magnetocapacity sign have been observed in room temperature in the paramagnetic region. A correlation of the changes in the dielectric permittivity and the magnetic susceptibility with temperature has been revealed. The magnetocapacity is described using the model with orbital electron ordering and the Maxwell–Wagner model.

1. INTRODUCTION

In materials in which a correlation between the magnetic and the electrical properties is observed [1, 2], magnetoelectrics and multiferroics [3, 4] are interesting both from fundamental and practical standpoints. Of a particular attention are materials that demonstrate magnetoelectric properties at room and higher temperatures due to their practical application in microelectronics for data recording and storage. Among such studied materials is bismuth ferrite $BiFeO_3$ [5, 6]. The giant magnetocapacity effect was observed in $LuFe_2O_4$ [7] at room temperature and was explained by fluctuations of charges with different values of spins in Fe^{2+} and Fe^{3+} ions as a result of splitting between two types of the charge order by an external magnetic field. The dependence of the orbital magnetic moments on polar distortions induced by electric field gives an "ion–orbital" contribution to the magnetoelectric response [8].

The systems with orbital ordering are characterized by anisotropic distribution of the electron density in a lattice with different hopping integral parameters [9, 10] and, correspondingly, with anisotropy of the current carrier mobility. Because of this, the migration electron polarization is also anisotropic. In the case when electrons have an orbital magnetic moment, for example, in the t_{2g} state, the anisotropy of the dielectric permittivity can be controlled by a magnetic field.

In electrically heterogeneous systems, the Maxwell–Wagner effect [11] and the contact effects can lead to giant values of the dielectric permittivity and the dielectric relaxation in the absence of the dipole relaxation [12]. The Maxwell–Wagner effect can also induce the magnetocapacity in the absence of the interaction between the magnetic and electrical subsystems under conditions of the magnetoresistance in a material [13]. These effects clearly demonstrate that the existence of the megnetocapacity is insufficient for these compounds to be referred to multiferroics. On the other hand, the magnetocapacity without magnetoelectric coupling can be more pragmatic for technological applications, because the existence of a long-range magnetic order is not necessary.

An electrically heterogeneous system with orbital degeneration can be obtained by the substitution of trivalent gadolinium ions for bivalent manganese ions. For example, the initial compounds, namely, manganese selenide and gadolinium selenide belong to semiconductors and degenerate semimetals, respectively, the electrical resistances of which differ by factors of 10^6-10^9 [14]. Gadolinium selenide and manganese selenide have an antiferromagnetic structure of the second-type ordering with the Néel temperatures $T_N = 63$ K [15] and $T_N = 137$ K [16], respectively. The spin-orbital and the Jahn–Teller interactions split t_{2g} electronic states and induce the spin splitting of the

spectrum of electron excitations. As a result, the dielectric properties can be controlled by electric and magnetic fields.

The aim of this work is to determine the contribution of the orbital ordering of electrons and the Maxwell–Wagner effect in the spin-disordered region in $Gd_xMn_{1-x}Se$ solid solutions to the magnetocapacity.

2. EXPERIMENTAL RESULTS AND DISCUSSION

The $Gd_xMn_{1-x}Se$ solid solutions were synthesized by the solid-phase reaction method described in [17] from powders of the initial compounds in evacuated quartz ampoules in a single-zone resistance furnace. The phase composition and the crystal structure of the $Gd_xMn_{1-x}Se$ samples were studied at 300 K using a DRON-3 diffractometer with CuK_α radiation. The X-ray diffraction data showed that the synthesized compounds have a NaCl-type face-centered cubic (fcc) structure typical of manganese monoselenide. As the degree of cation substitution (X) increases, unit cell parameter a increases from $a = 0.5440$ nm for MnSe to $a = 0.5520$ nm for $Gd_{0.2}Mn_{0.8}Se$ [17].

The capacity and the dielectric loss tangent ($\tan\delta$) were measured using an AM-3028 component analyzer in the temperature range 90–450 K in a zero magnetic field and in a magnetic field of $H = 8$ kOe. The magnetic field was applied parallel to the flat capacitor planes. The magnetocapacity effect $\delta\varepsilon_H = (Re(\varepsilon(H, T) - Re\varepsilon(0, T)))/Re(\varepsilon(0, T))$ is determined as a result of the studies of the complex dielectric permittivity. The spectral and temperature dependences of the dielectric constants can be used to the observation of the dipole electric moment and the determination of its characteristics, even when we are dealing with a local dipole moment in small clusters without a long-range order. The dielectric properties also give information of the charge transport and the processes of the charge ordering. The response of the dielectric properties on the action of a magnetic field gives a possibility of determining the main mechanisms that determine the correlation of the dielectric and the magnetic properties.

Figure 1 shows the temperature dependences of the real $Re(\varepsilon)$ and imaginary $Im(\varepsilon) = \tan\delta Re(\varepsilon)$ parts of the dielectric permittivity of the $Gd_{0.05}Mn_{0.95}Se$ sample. In the temperature range 220–330 K, the real part of the dielectric permittivity is doubled and shifts to higher temperatures with the frequency. The temperature of the dielectric loss maximum increases with frequency and is described by a linear function of the logarithm of frequency. The frequency dependence of the complex dielectric permittivity does not correspond to the Debye and Cool–Cool models that describe the dielectric properties of homogeneous systems with the exponential dependence of the relaxation of the polarization parameter. The dielectric relaxation frequency

corresponding to the maximum of $Im(\varepsilon)$ has an activation character and is well described by function $\ln\omega = A\exp(-\Delta E/kT)$ (Fig. 2b), where activation energy $\Delta E = 0.051$ eV and the exponent obeys the condition $\exp(-\Delta E/kT) \ll 1$ at $T < 350$ K. Below this temperature, we represent the frequency and the relaxation time (τ) in the forms:

$$\omega = B\exp\left(\exp\left(-\frac{\Delta E}{k_B T}\right)\right) = B\left(1 + \exp\left(-\frac{\Delta E}{k_B T}\right)\right);$$

$$\tau = \frac{\tau_0}{1 + \exp\left(-\frac{\Delta E}{k_B T}\right)}. \tag{1}$$

In Eqs. (1), the activation energy corresponds to the polaron energy (ε_p) with respect to the chemical potential (μ), $-\Delta E = \varepsilon_p - \mu < 0$. The relaxation time is proportional to the polaron density $\tau \propto N_p$. If the polaron energy is higher than the chemical potential $\varepsilon_p > \mu$, this hole-type polaron $N_{ph} = A/(1 + \exp(-\Delta E/k_B T))$. The imaginary component of the dielectric permittivity provided by scattering of polarons on optical phonons is proportional to $Im(\varepsilon)_{max} \propto (N_{pe} + N_{ph})$. The dielectric loss maxima increase with frequency and temperature in a zero magnetic field and smoothly decrease in magnetic field (Fig. 2b) as a result of the transformation of the electronic structure in magnetic field. For example, the spectral weight of polaron with hole-type conduction with ratio $N_{ph}/N_{pe} = 5.5$ dominates in a zero field and with electron-type conduction with $N_{pe}/N_{ph} = 2.3$ in magnetic field, which qualitatively describes the experimental results (Fig. 2b).

The temperature at which the dielectric permittivity increases sharply and the dielectric loss maximum is observed shifts to higher temperatures in a magnetic field. The activation energy increases by 5% in a magnetic field of 8 kOe. Figure 2a shows magnetocapacity $\delta\varepsilon_H = (Re(\varepsilon(H, T) - Re\varepsilon(0, T)))/Re(\varepsilon(0, T))$ for the composition with $x = 0.05$. On heating, the magnetocapacity changes its sign and decreases as the frequency increases.

The dispersion of nonuniform electronic states and local magnetic fields increases with the gadolinium ion concentration. The temperature range of the dielectric loss increases in the composition with $x = 0.2$ (Fig. 3); the temperatures of the maxima of the imaginary part of the dielectric permittivity do not almost shift in magnetic field, and the dielectric loss increases (Fig. 3). In magnetic field, the dielectric permittivity increases sharper (Fig. 3); as a result, the magnetocapacity changes its sign from negative values (5–7)% to positive (4–5)% in the temperature range (140–400) K (Fig. 4a).

The temperatures of the maxima of the dielectric loss increase within 1% in magnetic field and increase with frequency. The relaxation frequency is described
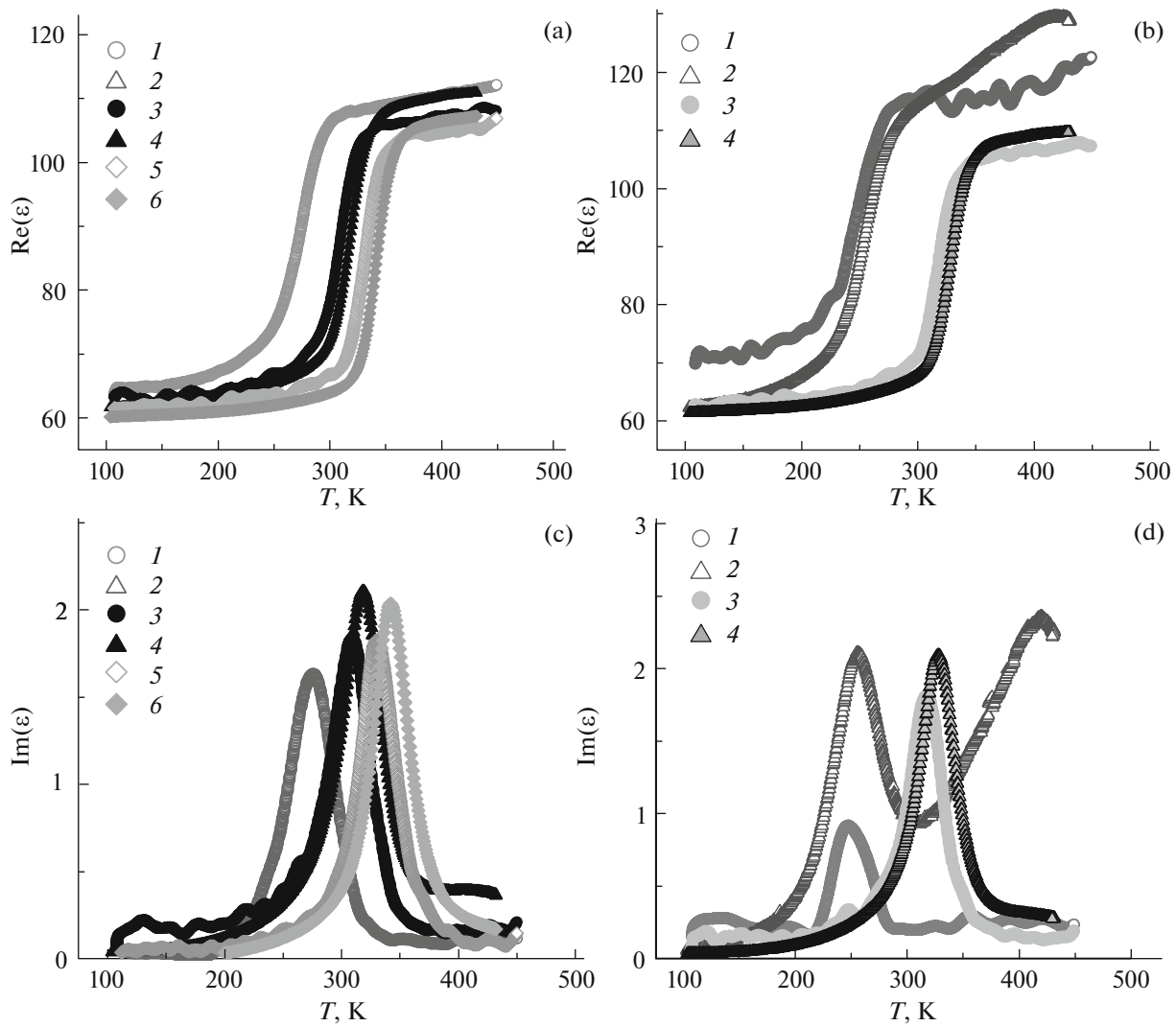
Fig. 1. (a, b) Real Re(ε) and (c, d) imaginary Im(ε) components of the dielectric permittivity of the $Gd_{0.05}Mn_{0.95}Se$ sample in magnetic field $H = (1, 3, 5)$ 0 and $(2, 4, 6)$ 8 kOe measured at frequencies ω = (a, c) 5, 50, and 300 kHz and (b, d) 1, 100 kHz vs. temperature.

by exponential dependence (1) with activation energy $\Delta E = 0.035$ eV. The dielectric loss is related to scattering of hole-type polarons, and function $Im(ε)_{max} = AN_{ph}$ adequately describes the experimental data (Fig. 4b).

The change in the dielectric permittivity with temperature is accompanied by a deviation of the magnetic susceptibility from the Curie−Weiss law. Figure 5 shows the temperature dependence of the inverse magnetic susceptibility. At temperatures higher than $T = 250$ K, the inverse magnetic susceptibility is well described by the Curie−Weiss law with the paramagnetic temperature Θ that has a negative value and increases from $\Theta_P = -350$ K for MnSe to $\Theta_P = -98$ K for $x = 0.2$, which demonstrates the formation of the ferromagnetic exchange as gadolinium is substituted

for manganese. This decrease is due to the localization of electrons in clusters with gadolinium ions and the formation of the ferromagnetic exchange interaction at the Mn−Gd interface as a result of the kinetic exchange interaction.

The effective magnetic moment increases with the concentration from $5.9\mu_B$ for MnSe to $6.39\mu_B$ in $Mn_{0.8}Gd_{0.2}Se$ (Fig. 6a). In this case, the magnetic moment is $(1-3)\%$ larger than the sum of the effective magnetic moments of manganese and gadolinium ions $\mu^{ef}(x) = (1-x)\mu_{Mn}^{ef} + x\mu_{Gd}^{ef}$. The splitting of the t_{2g} electron states leads to a decrease in the local symmetry of manganese ion clusters and a changes in the g factor and the effective magnetic moment $\mu^{ef} = g\sqrt{(S(S+1))}\mu_B$. The concentration of these clusters
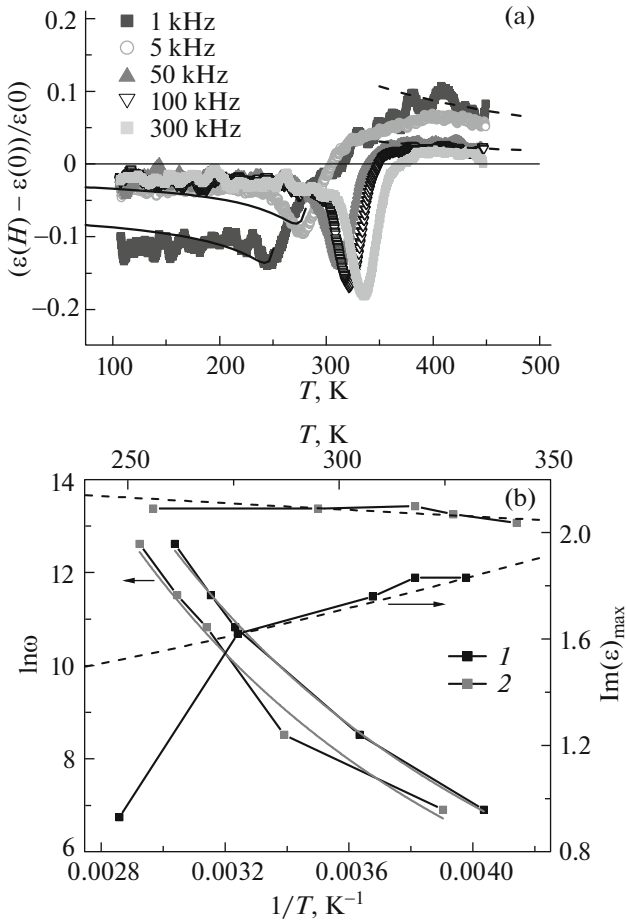
Fig. 2. (a) Magnetocapacity $\delta\varepsilon_H$ measured as a function of temperature at frequencies $\omega = 1, 5, 50, 100,$ and 300 kHz in magnetic field $H = 8$ kOe for the composition with $x = 0.05$. The theoretical value of the magnetocapacity in (solid line) the model with orbital ordering (3) and (dashed line) in the Maxwell–Wagner model (4) with $\delta\varepsilon \sim 1/T^{3/2}$. (b) Logarithm of the relaxation frequency vs. reciprocal temperature (1) without field and (2) in magnetic field $H = 8$ kOe; (1) the fitting function (the left axis). The maximum of $\mathrm{Im}(\varepsilon)$ vs. the temperature. (dashed line) Adjustable functions $\mathrm{Im}(\varepsilon)_{max} = AN_{pe} + BN_{ph}$ (the right axis).

containing gadolinium ions surrounded with manganese ions with magnetic moments μ_{Mn}^{ef*} is $c_g = zx(1 - x)^{z-1}$, and the change of the effective magnetic moment as a function of the concentration has the form $\mu^{ef}(x)/\mu_{Mn}^{ef} = 1 - x - (1 - \lambda)c_g + \gamma x$, where $\lambda = \mu_{Mn}^{ef*}/\mu_{Mn}^{ef}$ and $\gamma = \mu_{Gd}^{ef}/\mu_{Mn}^{ef} = 1.34$. The fitting function describes the experimental results with parameter $\lambda = 1.055$, which corresponds to the increase in g-factor to 2.11. In a disordered system, the orbital magnetic moment that interacts to the spin moment is induced as a result of the spin–orbital interaction.

3. MODEL

According to the LSDA + U theoretical calculations [18], trivalent gadolinium selenide is a metal, in which the conduction band is due to $5d$ electrons and the upper Hubbard subband of $4f$ electrons is 2 eV higher than the Fermi level. The conduction band width is 8 eV. The substitution of trivalent gadolinium ions for bivalent manganese leads to an electron doping and degeneration of t_{2g} electron states that is split by the Jahn–Teller and spin–orbit interactions. In MnSe, the current carriers are holes that interact with optical vibrational modes and form lattice polarons that are pinned at the Mn–Gd interface. The pinning of the polarons reduces the local symmetry and induces an orbital magnetic moment on the site. It is found that the dielectric permittivity of manganites decreases as a result of condensation of one of the components of the triply degenerate T_{1g} vibrational mode in the center and at the boundaries of the Brillouin zone [19]. This mechanism leads to a change in the dielectric permittivity in the $Mn_{1-x}Gd_xSe$ solid solution.

Estimate the spin-orbit coupling constant (α) for an ion in an octahedral approximation from the relationship $\alpha = 2(1 - g/\Delta)$, where Δ is the splitting of the t_{2g} states that varies within $\Delta = 0.1-0.15$ eV and $\alpha \sim 10$ meV in $3d$ metals with the perovskite structure. The uniaxial magnetic anisotropy constant is $D = \alpha^2/\Delta \sim 10^{-3}$ eV, and the anisotropy field is an order lower than the exchange field: $H_A/H_{ex} = D/zJ \sim 0.1$. The appearance of the longitudinal component of the angular magnetic moment is accompanied by the magnetic and dielectric anisotropies in the plane; it follows from the anisotropic distribution of the electron density of the electron wave functions. We use a simple relationship between the components of the dielectric permittivity $\varepsilon_{xx} > \varepsilon_{yy}$ for the projections of the orbital magnetic moment $L^z = +1$, $\varepsilon_{xx} < \varepsilon_{yy}$ for $L_z = -1$, and $\varepsilon_x = \varepsilon_{yy}$ for $L_z = 0$.

At low concentrations $x < x_c$ ($x_c = 0.17$ is the percolation concentration), the main contribution to the magnetic and dielectric properties is given by clusters of gadolinium ions that have only manganese ions in their nearest environments. Below 250 K, a local deformation of the lattice and the freezing of orbital magnetic moments appear in the vicinity of these clusters with a random anisotropy of the dielectric permittivity. The nanoregions have a short-range ferromagnetic order, and we describe the change in the orientation of dipole moments (M_L) clusters in magnetic field H in a superparamagnet model.

Represent the energy of superparamagnetic particles as $W = M_L H\cos\theta + M_L H_A\cos(\gamma - \theta)$, where angle θ gives the magnetic moment direction with respect to the magnetic field, γ is the angle between the magnetic
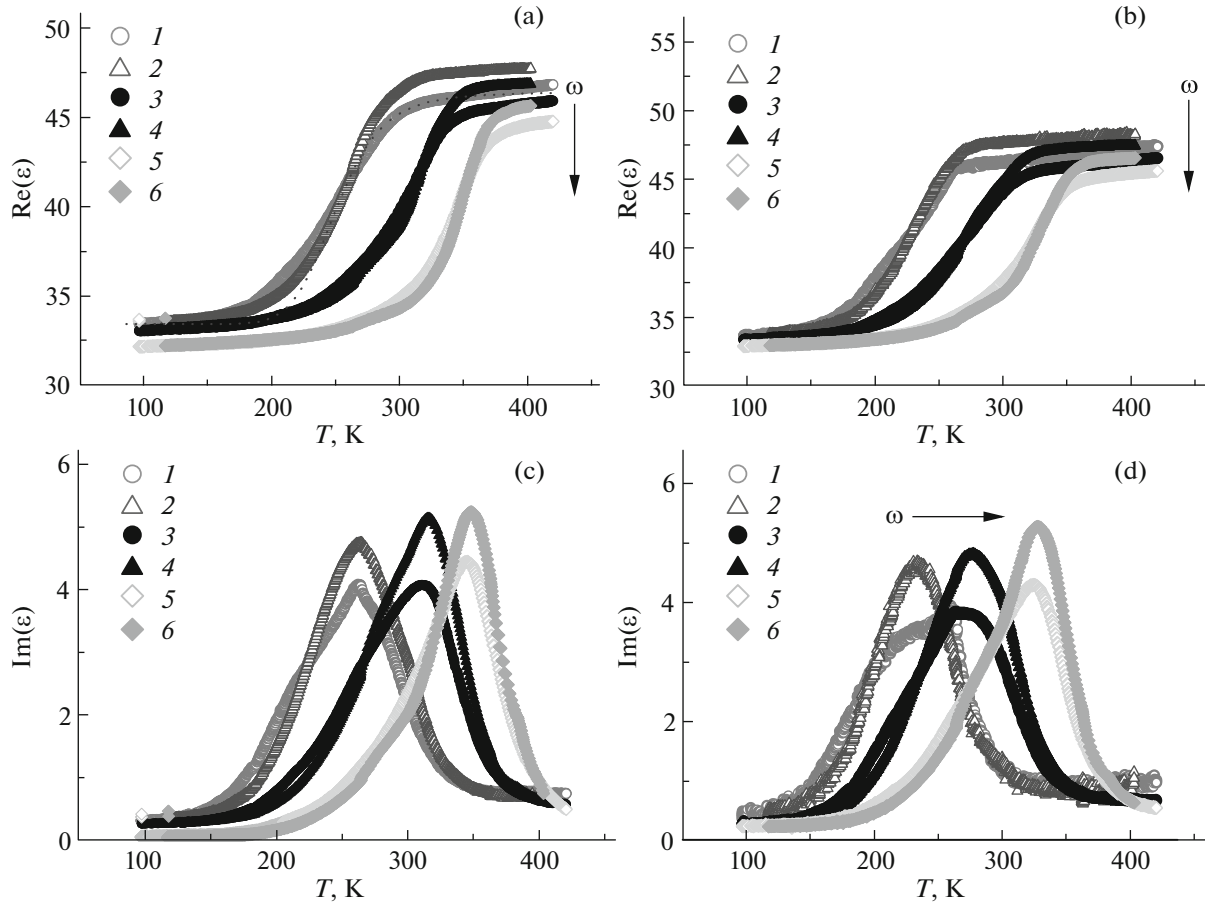
Fig. 3. (a, b) Real Re(ε) and (c, d) imaginary parts of the dielectric permittivity of the $Gd_{0.2}Mn_{0.8}Se$ measured as functions of temperature in (*1, 2, 3*) a zero magnetic field and (*2, 4, 6*) in magnetic field $H = 8$ kOe at frequencies (a, c) ω = 5, 50, and 300 kHz and (b, d) ω = 1, 10, 100 kHz.

field and the anisotropy field, and H_A is the anisotropy field. The equilibrium direction of the orbital moment is determined by the relationship $\tan\theta = H_A\sin\gamma/(H + H_A\cos\gamma)$. We represent the change in the orbital moment of all particles in magnetic field as

$$M_L^0\Delta\theta = M_L^0\left(\langle\cos\theta\rangle_H - \langle\cos\theta\rangle_0\right)$$

$$= M_L^0\left(\sum_i \frac{1}{\sqrt{1 + \dfrac{H_A^2\sin^2\gamma_i}{(H + H_A\cos\gamma_i)}}} - \frac{1}{1 + \tan^2\gamma_i}\right). \quad (2)$$

The relative change in the dielectric permittivity in magnetic field is proportional to the dielectric permittivity anisotropy $\Delta\varepsilon = \varepsilon_{xx} - \varepsilon_{yy}$ and to the change in the orbital moment in magnetic field

$$\delta\varepsilon = \Delta\varepsilon(M_L(H) - M_L(0)) = \Delta\varepsilon m_L(1 - T/T_c)^\beta$$

$$\times\left(\sum_i \frac{1}{\sqrt{1 + \sin^2\gamma_i\left[\dfrac{\lambda}{(1 - T/T_{c,B})^2} + \cos\gamma_i\right]^2}}\right. \quad (3)$$

$$\left. - \frac{1}{\sqrt{1 + \tan^2\gamma_i}}\right),$$

where $\lambda = H/H_A$. In the medium field approximation, the magnetization in the transition region has the form $M = m_L(1 - T/T_C)^\beta$, where $\beta = 0.5$. The anisotropy field also has a power dependence $H_A = D(1 - $
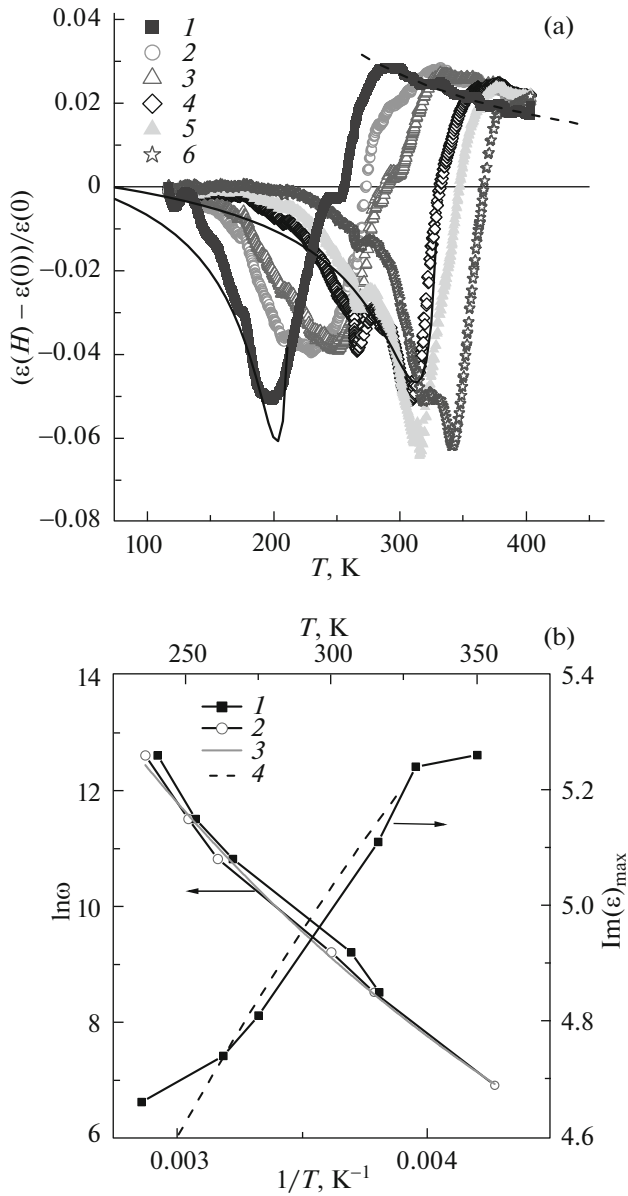
Fig. 4. (a) Magnetocapacity $\delta\varepsilon_H$ measured as a function of temperature at frequencies ω = (*1*) 1, (*2*) 5, (*3*) 10, (*4*) 50, (*5*) 100, and (*6*) 300 kHz in magnetic field H = 8 kOe for the composition with x = 0.2. The theoretical value of the magnetocapacity in $Gd_{0.2}Mn_{0.8}Se$ (solid line) the model with orbital ordering (*3*) and (dashed line) in the Maxwell−Wagner model (*4*) with $\delta\varepsilon \sim 1/T^{3/2}$. (b) Logarithm of the relaxation frequency vs. reciprocal temperature measured (*1*) in a zero magnetic field and (*2*) in magnetic field H = 8 kOe; the fitting function (1) (curve *3*) (the left axis). The maximum of $Im(\varepsilon)_{max}$ vs. the temperature and adjustable functions $Im(\varepsilon)_{max} = AN_{ph}$ (*4*) (the right axis).

$T/T_{c, B})^n$. In the case x = 0.05, the experimental data on the magnetocapacity measured at frequency ω = 1 kHz (Fig. 2) are adequately described by function (3) with the following parameters: orbital moment of a
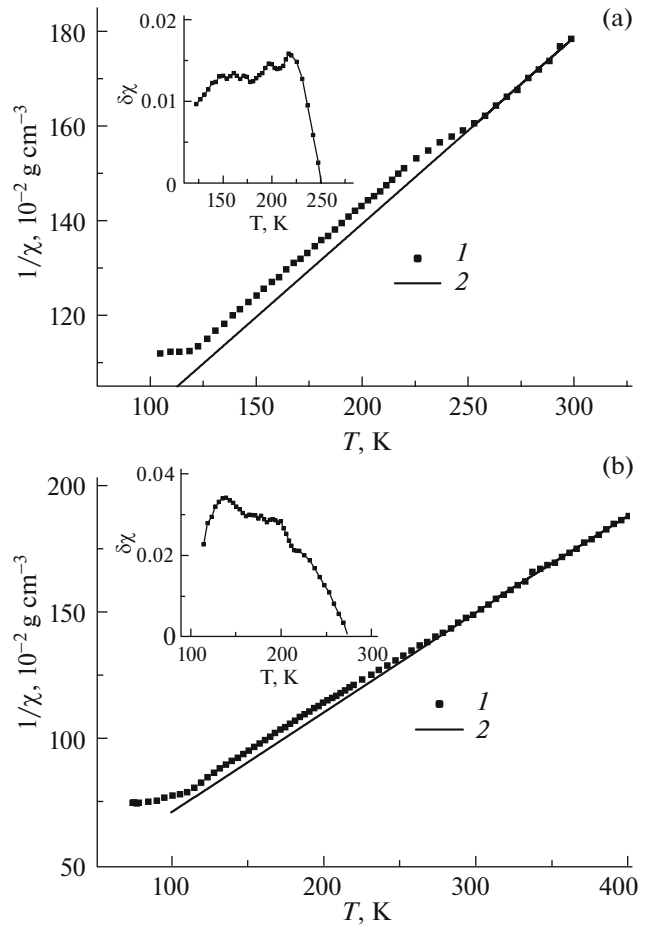

Fig. 5. Temperature dependences of the inverse susceptibilities for two compositions x = (a) 0.05 and (b) 0.2. The inserts show the difference between (*2*) Curie−Weiss magnetic susceptibilities and (*1*) the experimental values $\delta\varepsilon$ = $(\chi^{CW} - \chi^{EX})/\chi^{EX}$ as functions of temperature.

cluster m_L = 0.1, λ = 0.01, n = 1.5, and the polaron pinning temperature with the formation of the orbital magnetic moment $T_{c, B}$ = 252 K. As the frequency increases, the pinning temperature of lattice polarons at the Mn−Gd interface increases and the minima in dependence $\delta\varepsilon(T)$ are qualitatively described by function (3). In going to the band-type conduction, the current carrier mobility increases and the magnetocapacity is due to the Maxwell−Wagner mechanism, when the heterogeneity mixes real and imaginary modes of the dielectric response, longitudinal and transverse vibrational modes. The heterogeneity also causes the dc magnetoresistance as a result of mixing the Hall resistance with the longitudinal resistance, providing linear field dependence. In the 2D model of two-component strongly heterogeneous medium with the conductivity σ_1 = 0, $\varepsilon_1 = \varepsilon$ and $\sigma_2 = \sigma$, and ε_2 = 0, the exact expression for the longitudinal dielectric
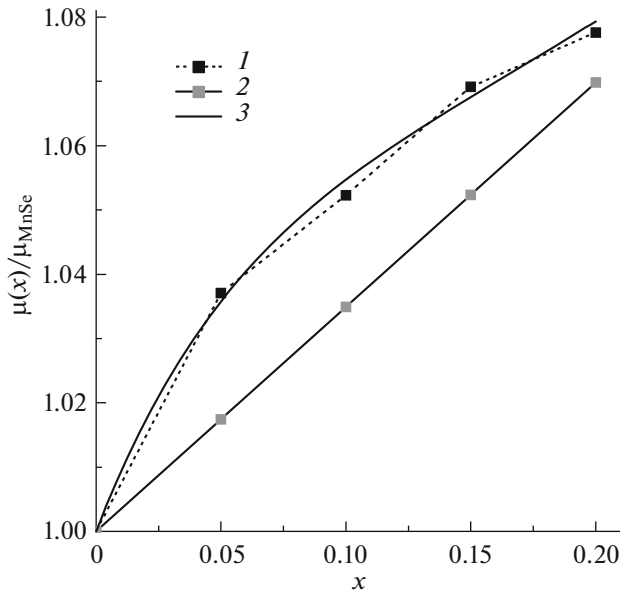
Fig. 6. Dependence of the magnetic moment of cations $\mu^{ef}(x)/\mu_{Mn}^{ef}$ on the gadolinium ion concentration in the Gd_xMn_{1-x} Se solid solution: (*1*) experiment, (*2*) the sum of the effective moments $\mu^{ef}(x)/\mu_{Mn}^{ef} = 1 - x + x\mu_{Gd}^{ef}/\mu_{Mn}^{ef}$, and (*3*) model calculations of $\mu^{ef}(x)/\mu_{Mn}^{ef} = 1 - x - (1-\lambda)c_g + 1.34x$, where $\lambda = \mu_{Mn}^{ef*}/\mu_{Mn}^{ef}$.

permittivity in the case of the equal concentrations is [13]

$$\varepsilon_{xx}(\omega) = \varepsilon \frac{(1 + i\omega\tau)}{\sqrt{i\omega\tau}\sqrt{(1+i\omega\tau)^2 - (\omega\tau\beta)^2}}, \qquad (4)$$

where $\beta = \mu H$, where μ is the charge carrier mobility in magnetic field. The real and imaginary components of the dielectric permittivity increase in magnetic field, and the dependence of the dielectric permittivity on the mobility and magnetic field is described by the power function $\varepsilon_{xx}(\beta)/\varepsilon = A\beta^{0.7}$ at $\omega\tau\beta < 1$ and $\omega\tau \approx 1$. In this model, we estimated the carrier mobility from the magnetocapacity $\delta\varepsilon = (\varepsilon_{xx}(\beta) - \varepsilon_{xx}(0))/\varepsilon(0) = 0.6(\mu H)^{0.7}$ and obtained $\mu \approx 0.01 T^{-1}$ at the frequency $\omega = 1$ kHz. At temperatures higher than 250 K, the short-range orbital order is retained and the magnetocapacity is due to two competing mechanisms: a decrease in the dielectric permittivity in magnetic field as a result of enhancing of correlations between orbital magnetic moments and an increase in ε due to the Maxwell–Wagner effect. The charge carrier mobility decreases at high temperatures $T > 350$ K due to scattering by phonons, according to the power function $\mu = A/T^{3/2}$. The magnetocapacity provided by a change in the mobility adequately describes the experimental results (Fig. 2).

At concentrations higher than the percolation concentration ($x > x_c$), a long-range orbital order with a nonzero resulting orbital moments forms at a Mn–Gd interface. The main contribution to the magnetocapacity is due to the dielectric permittivity anisotropy controlled by magnetic field due to spin–orbit coupling in the vicinity of a Gd–Mn interface. The experimental data are adequately described by function (3) with parameters $\beta = 0.33$, $n = 2$, and $M_L(0) = 0.025$. At higher temperatures $T > 250$ K, the magnetocapacity is caused by a decrease in the current carrier mobility $\delta\varepsilon(T) = A/T^{3/2}$ as a result of their scattering by phonons.

4. CONCLUSIONS

The magnetocapacity effect has been revealed in the $Gd_xMn_{1-x}S$ solid solutions at temperatures several times higher than the Néel temperature with the change in sign with temperature. Logarithm of the frequency of the dielectric loss relaxation exponentially increases on heating and is dependent on magnetic field. It was found that the effective magnetic moment nonlinearly increases with the concentration due to the contribution of the orbital magnetic moment of manganese ions near the Mn–Gd interface. We revealed the correlation between the temperatures, at which the magnetic susceptibility increases and the dielectric permittivity decreases.

The experimental data are explained in the model with the orbital ordering. At temperatures lower than the Debye temperature, polarons are pinned at the interface with the appearance of the orbital magnetic moment at a site and the dielectric permittivity anisotropy. At gadolinium ion concentrations lower than the percolation concentration, the orbital "glass" forms and, at higher concentrations, a long-range orbital order is induced at the interface. The change in the orbital correlations of the magnetic angular moment in magnetic field changes the dielectric permittivity anisotropy. The delocalization of electrons and the transition to the band-type conduction leads to the positive magnetocapacity as a result of the Maxwell–Wagner.

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