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Magnetoresistance on direct and alternating current in manganese selenide substituted with thulium

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The scale of electrical inhomogeneity in manganese selenide substituted with thulium for magnetoresistance at direct and alternating current is investigated using I-V characteristics measured without a field and in a magnetic field in the paramagnetic phase. A correlation was found between the nonlinearity of the I-V characteristic and the magnetoresistance at direct current. A decrease in resistance in a magnetic field on alternating current was discovered with increasing concentration of thulium ions. The difference in magnetoresistance for direct and alternating current is explained by taking into account the contribution of dielectric constant in the magnetodielectric resonance model.

Keywords: semiconductors, selenides, magnetoresistance, nonlinearity of the current-voltage characteristic.

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1. Introduction

Semiconductors with a magnetoresistive effect are used widely in practice; from a theoretical standpoint, it is of interest to investigate the mechanism of influence of the magnetic field on semiconductors in the paramagnetic state [1–3]. For example, the change in magnetoresistance (MR) sign with the applied field in zinc oxide films is attributed to the competing contributions of the $s-d$ interaction in the multiband model (conduction band and impurity band) [4–6]. In topological insulators Bi_2Te_3 , the magnetoresistance remains approximately at 400% within a wide temperature range of 3–300 K and reaches its maximum of 600% at 340 K without any signs of MR saturation in the magnetic field, which is attributable to the spin-orbit interaction [7–9]. In $\text{Ag}_{2\pm\delta}\text{Se}$ and $\text{Ag}_{2\pm\delta}\text{Te}$ semiconductors, the magnetoresistance is explained by the nonuniformity [10] and dispersion in the carrier mobility distribution function, which leads to Hall conductivity from impurity centers [11–14]. Multiple scattering of electrons in low-mobility regions in inhomogeneous semiconductors results in induced disorder, where free carriers are frozen in high-mobility channels at low temperatures. The gapless linear law of dispersion is not a prerequisite for the emergence of disorder-induced magnetoresistance [15]. This disorder-induced magnetoresistance depends strongly on temperature, has a nonmonotonic temperature dependence, and is observed at high temperatures.

The electrical inhomogeneity in manganese chalcogenides may be adjusted by substituting manganese ions with rare-earth ions with variable valence [16–18]. The valence may change under chemical pressure during heating, which is accompanied by a change in the scattering potential and

carrier mobility [19]. Nonstoichiometry may induce an additional charge transfer channel in which current carriers are frozen at low temperatures. The valence of thulium ions changes from a trivalent state in TmS [20] to a divalent state in TmTe [21] via an intermediate valence in TmSe [22]. Manganese selenide substituted with thulium is a promising material for the examination of magnetoresistive effects.

In such systems, magnetoresistive effects are governed by the electrical inhomogeneity and carrier mobility, which depends on the measurement time. The scale of electrical inhomogeneity in a system with migration polarization will manifest itself differently in DC and AC magnetoresistance; the present study is focused on the detection of these effects.

2. DC magnetoresistance

Solid solutions of manganese selenide substituted with thulium were prepared with account for nonstoichiometry in cations in order to raise the valence of thulium ions. The $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$, $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ samples were synthesized by solid-state reaction in evacuated quartz vessels. The synthesis technique was detailed in [23]. The results of X-ray diffraction analysis of selenides verify that the face-centered cubic lattice of the NaCl type is preserved after substitution. The DC magnetoresistance is determined from current-voltage characteristics (CVCs) measured in zero field and in a certain magnetic field. Figure 1 shows the CVCs for two samples.

The dependence of current on voltage is nonlinear. We characterize the deviation from the Ohm's law by the relative difference in resistance at the maximum and minimum (in magnitude) voltages $(R(U_{\min}) - R(U_{\max}))/R(U_{\max})$, which is shown in Figure 1, *d*. The CVC nonlinearity

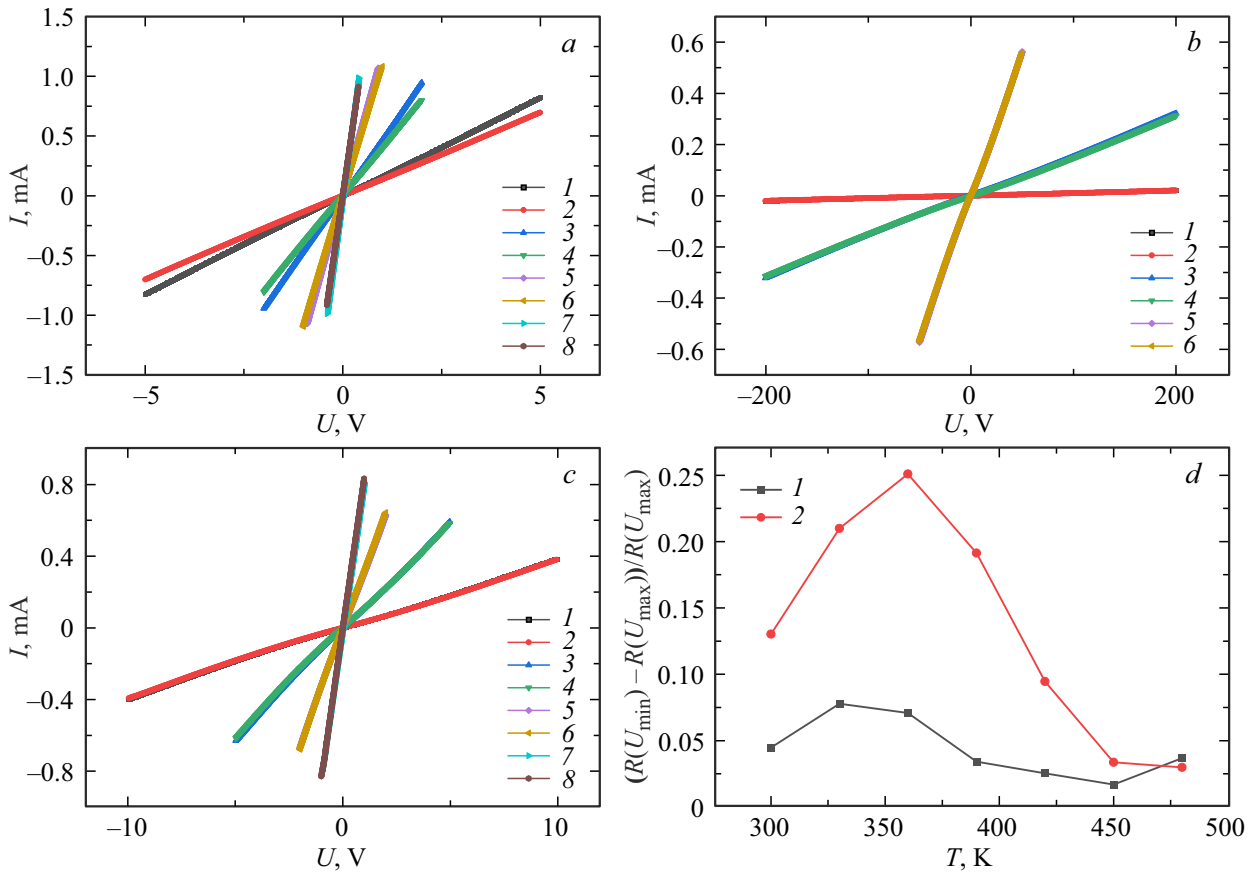


Figure 1. $I(V)$ characteristic: *a* — at $T = 390$ K (curves 1, 2), 420 (3, 4), 450 (5, 6), 480 (7, 8) for sample $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$; *b* — at $T = 300$ K (1, 2), 330 (3, 4), 360 (5, 6) for $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$; *c* — at $T = 390$ K (1, 2), 420 (3, 4), 450 (5, 6), 480 (7, 8) for $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$; in zero field (1, 3, 5, 7) and in magnetic field $H = 8$ kOe (2, 4, 6, 8). *d* — Temperature dependence of $(R(U_{\min}) - R(U_{\max})) / R(U_{\max})$ for samples $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (1) and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (2).

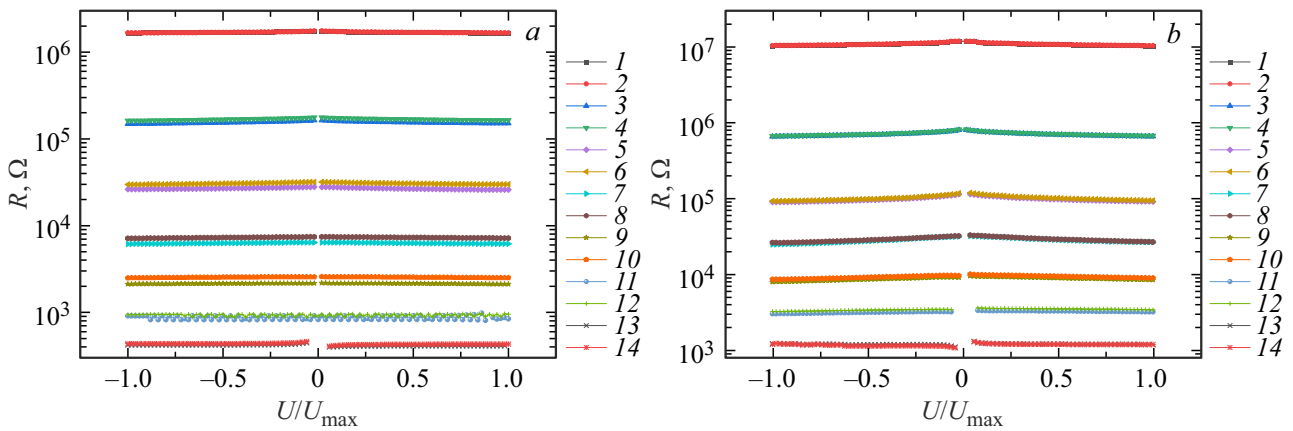


Figure 2. Electrical resistance versus normalized voltage U/U_{\max} for samples $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (*a*) and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (*b*) measured in zero field and in magnetic field $H = 8$ kOe at temperatures $T = 300$ K (curves 1, 2), 330 (3, 4), 360 (5, 6), 390 (7, 8), 420 (9, 10), 450 (11, 12), 480 (13, 14); in zero field (1, 3, 5, 7, 9, 11, 13) and in magnetic field $H = 8$ kOe (2, 4, 6, 8, 10, 12, 14).

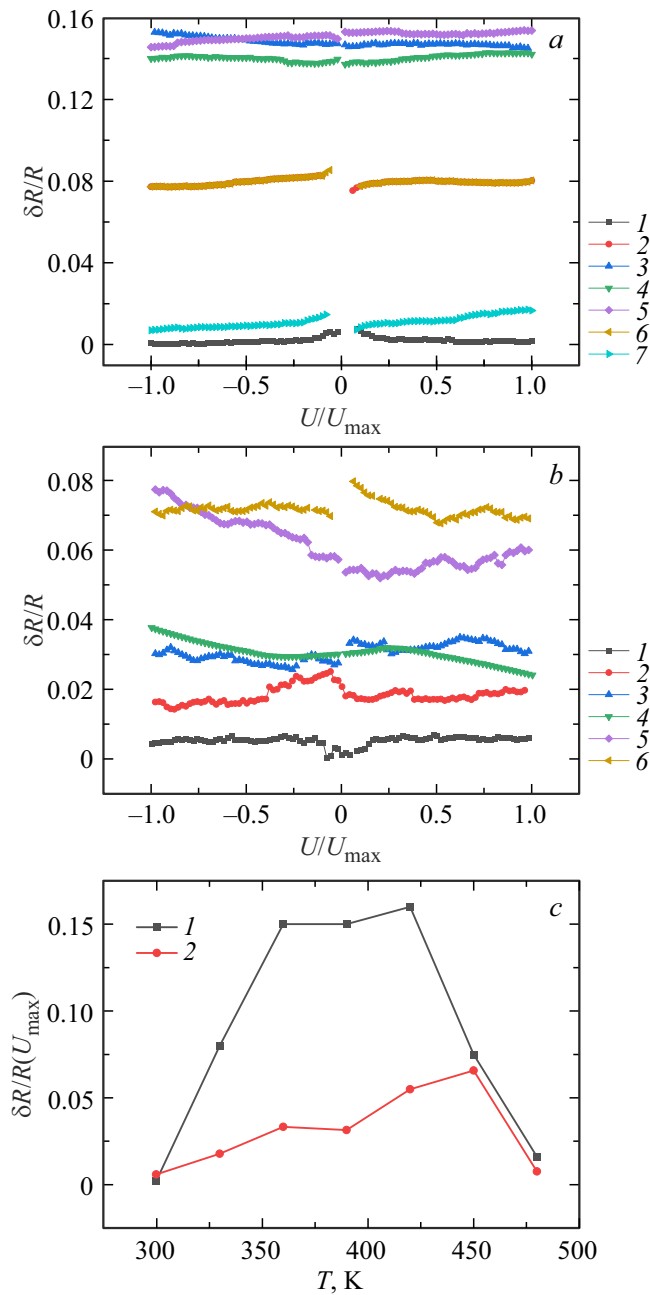


Figure 3. Dependences of magnetoresistance on normalized voltage U/U_{\max} for samples $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (a) and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (b) at temperatures $T = 300, 330, 360, 390, 420, 450,$ and 480 K — curves 1–7, respectively; c — temperature dependence of $\delta R/R(U_{\max})$ for $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (1) and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (2).

increases under heating and goes through a maximum in the temperature interval of 340–360 K.

The resistance decreases exponentially under heating, while the current rises sharply; therefore, we set the upper current limit at 1 mA. Figure 2 shows the dependences of resistance on normalized voltage measured in zero field and in magnetic field $H = 8$ kOe directed perpendicular to the current.

The resistance change in the magnetic field is defined as $\delta R/R = (R(H) - R(0))/R(0)$ (see Figure 3). The magnitude of magnetoresistance is virtually independent of voltage. The magnetoresistance of the $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ sample at $T = 390$ and 420 K is asymmetric.

The magnetoresistance goes through a maximum under heating and is correlated with an increase in the CVC nonlinearity. The quadratic dependence of current on voltage (Mott law) stems from the presence of an uncompensated charge, which induces current via its electrostatic field. Power-law dependence $I = AU^n$ is established in semiconductors with donors and deep traps. The power-law growth is controlled by the impurity energy level [24] and the Frenkel effect (reduction of the ionization energy by an external electric field), which is screened by free charges [25]. The power-law stage ends when all traps and impurities get filled (as allowed by the thermal equilibrium) and is followed by the Mott–Gurney regime [26].

In a magnetic field, the radius of electrical inhomogeneity increases as a result of charge localization on traps near thulium ions, which results in an increase in electrical resistance in a magnetic field at direct current. Inhomogeneity plays a significant part in DC magnetotransport properties in disordered media, since the Hall resistance is mixed here with the longitudinal conductivity components, giving rise to a linear dependence of magnetoresistance on the magnetic field [27].

3. AC magnetoresistance

DC and AC conductivities differ fundamentally in semiconductors with electrical inhomogeneity. Conductivity is dependent on frequency ($\sigma = A\omega^s$) in semiconductors with hopping conduction [28]. In addition, one needs to take into account the complex nature of conductivity $\sigma(\omega, H)$, which assumes the following form in a transverse magnetic field:

$$\sigma(\omega, H) = \frac{\sigma}{1 + \beta^2} \begin{pmatrix} 1 & \beta \\ -\beta & 1 \end{pmatrix} + i\omega\varepsilon(\omega, H). \quad (1)$$

Here, σ is the DC conductivity scalar; ε is the dielectric permittivity, which also depends on frequency and magnetic field; and $\beta = \mu H$, where μ is the carrier mobility in a magnetic field.

The AC resistance is shown in Figure 4. At low frequencies ($\omega\tau < 1$), the resistance does not depend on frequency, since capacitive resistance is dominant. At high frequencies ($\omega\tau > 1$), the voltage drops mostly across a resistor, since there is not enough time to charge a capacitor.

The influence of the magnetic field on the AC conduction is established by an additional contribution due to the magnetocapacitive effect. Figure 5 shows the relative variation of AC resistance in magnetic field $H = 8$ kOe. In the $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ composition, the magnetoresistance changes sign above 10^5 Hz, and the conductivity for $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ above 360 K decreases in the magnetic field.

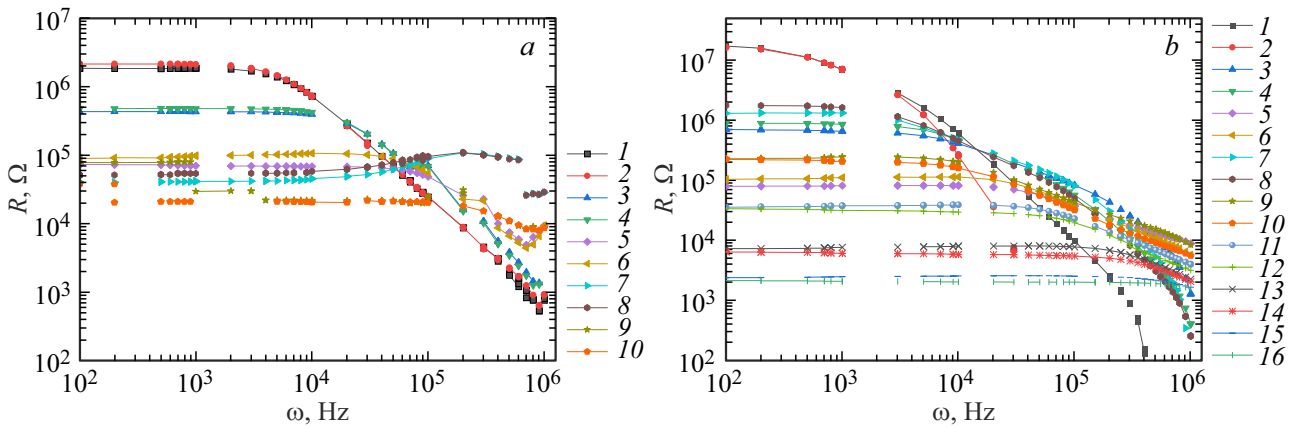


Figure 4. Frequency dependence of resistance for $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (a) at $T = 300$ K (curves 1, 2), 330 (3, 4), 360 (5, 6), 390 (7, 8), 420 (9, 10) and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (b) at $T = 300$ K (1, 2), 310 (3, 4), 320 (5, 6), 330 (7, 8), 360 (9, 10), 390 (11, 12), 420 (13, 14), 450 (15, 16); in zero field (1, 3, 5, 7, 9, 11, 13, 15) and in magnetic field $H = 8$ kOe (2, 4, 6, 8, 10, 12, 14, 16).

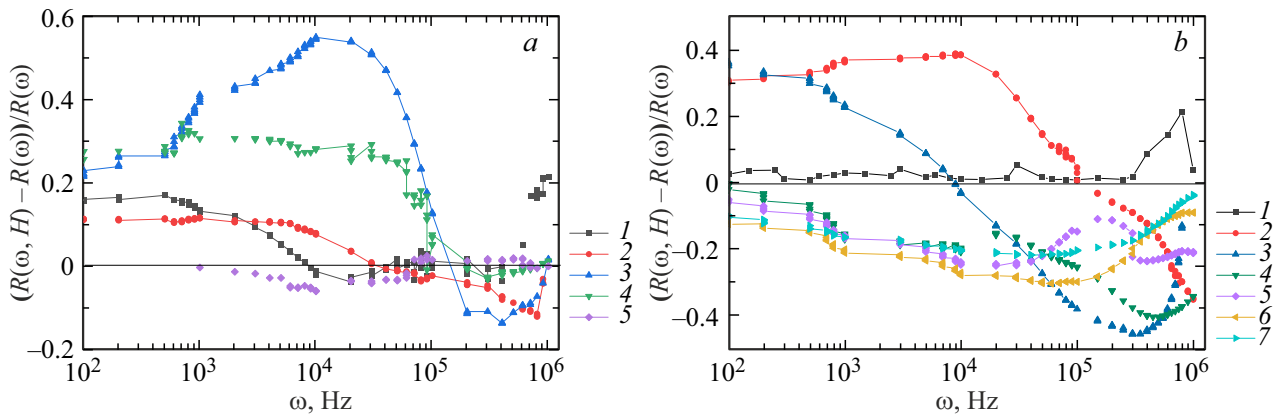


Figure 5. Frequency dependence of magnetoresistance for $\text{Tm}_{0.04}\text{Mn}_{0.95}\text{Se}$ (a) at $T = 300, 330, 360, 390,$ and 420 K — curves 1–5, respectively, and $\text{Tm}_{0.08}\text{Mn}_{0.9}\text{Se}$ (b) at $T = 300, 320, 330, 360, 390, 420,$ and 450 K — curves 1–7, respectively.

The change in magnetocapacitance of the samples is characterized qualitatively by the magnetoelectric resonance model. In a two-phase two-dimensional composite medium with limit parameters $\sigma_1 = 0$, $\varepsilon = \varepsilon_1$ with concentration x and $\sigma_2 = \sigma$, $\varepsilon = 0$ with $(1 - x)$, a numerical solution was found for the frequency dependence of the dielectric response in constant magnetic fields [29]. The permittivity was found to increase in a magnetic field in frequency range $\omega\tau = (0.04 - 0.2)$ with concentration $x = 0.5$. This increase is the result of mixing of the Hall resistance with the longitudinal conductivity components [30].

In a homogeneous system, $\varepsilon(\omega)$ decreases in a magnetic field. The presence of electrically inhomogeneous states leads to an increase in permittivity in a magnetic field [31]. Electrically inhomogeneous states exist above 330 K in the sample with $x = 0.1$ within the $10^{-2} - 10^{-6}$ s time interval.

4. Conclusion

Electrically inhomogeneous states were found by analyzing the CVCs of solid solutions of nonstoichiometric manganese selenide substituted with thulium. Their nonlinearity

goes through a maximum and is correlated in temperature with positive DC magnetoresistance. An increase in electrical inhomogeneity leads to a change in the sign of AC magnetoresistance with frequency and temperature variations. A reduction in electrical resistance in a magnetic field is associated with an increase in permittivity as a result of mixing of the Hall resistance with the longitudinal conductivity components.

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Conflict of interest

The authors declare that they have no conflict of interest.

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