UDC 537.312:538.911'956
Doi: 10.31772/2712-8970-2024-25-4-531-538

Для цитирования: Харьков А. М., Ситников М. Н., Аплеснин С. С. Магнитоимпеданс в тулий марганцевом халькогениде // Сибирский аэрокосмический журнал. 2024. Т. 25, № 4. С. 531–538. Doi: 10.31772/2712-8970-2024-25-4-531-538.

For citation: Kharkov A. M., Sitnikov M. N., Aplesnin S. S. [Magnetoimpedance in thulium manganese chalcogenide]. *Siberian Aerospace Journal.* 2024, Vol. 25, No. 4, P. 531–538. Doi: 10.31772/2712-8970-2024-25-4-531-538.

Магнитоимпеданс в тулий марганцевом халькогениде

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Управление транспортными характеристиками под действием магнитного поля является перспективным с точки зрения создания датчиков магнитного поля устойчивых к радиации. Исследуется импеданс и его компоненты в тулий марганцевом халькогениде в интервале частот 10^2–10^6 Гц. Найдена область температур с превалирующим вкладом реактивной и активной частей импеданса. Компоненты импеданса описываются в модели Дебая. При замещении марганца ионами тулия частоты максимумов мнимой компоненты импеданса смещаются в сторону высоких частот в селениде марганца на два порядка. С ростом концентрации замещения ионами тулия в селенидах найдено два времени релаксации по сравнению с сульфидами. Найден активационный характер времени релаксации, энергия активации от концентрации ионов тулия. Установлено увеличение импеданса в магнитном поле в области малых концентраций и смена знака импеданса по температуре для больших концентраций. Магнитоимпеданс в халькогенидах проходит через максимум при нагревании образцов. Увеличение импеданса в магнитном поле обусловлено изменением диагональной компоненты диэлектрической проницаемости в магнитном поле, которая пропорциональна проводимости Положительное значение магнитоимпеданса описывается в модели электрически неоднородной среды. Из импеданса можно получить информацию об электрической неоднородности материала.

Ключевые слова: полупроводники, магнитоимпеданс, модель Дебая.

Magnetoimpedance in thulium manganese chalcogenide

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Control of transport characteristics under the influence of a magnetic field is promising from the point of view of creating magnetic field sensors resistant to radiation. The impedance and its components in thulium manganese chalcogenide in the frequency range of 10^2–10^6 Hz are studied. The temperature range with a prevailing contribution of the reactive and active parts of the impedance is found. The impedance components are described in the Debye model. When manganese is replaced by thulium ions, the frequencies of the maxima of the imaginary component of the impedance shift toward high frequencies in manganese selenide by two orders of magnitude. With an increase in the concentration of substitution by thulium ions in selenides, two relaxation times are found, compared with sulfides. The activation nature of the

relaxation time, the activation energy from the concentration of thulium ions are found. An increase in impedance in a magnetic field in the region of low concentrations and a change in the sign of the impedance with temperature for high concentrations are established. Magnetoimpedance in chalcogenides passes through a maximum when heating the samples. The increase in impedance in a magnetic field is due to a change in the diagonal component of the permittivity in a magnetic field, which is proportional to the conductivity. A positive value of magnetoimpedance is described in the model of an electrically inhomogeneous medium. From the impedance, information can be obtained about the electrical inhomogeneity of the material.

Keywords: semiconductors, magnetoimpedance, Debye model.

1. Introduction

The control of transport characteristics in semiconductors under the action of an external magnetic field is of interest from both fundamental and practical points of view [1-4]. In semiconductors with inhomogeneous electric charge distribution, the transport characteristics depend on the degree of inhomogeneity [5-8]. In the spectrum of electronic excitations in the forbidden zone, a finite electron density at the chemopotential level is formed as a result of charge localisation. At weak doping, the electron wave function remains localised. As the substitution concentration increases, delocalised states are formed in the centre of the zone and at some critical value an infinite cluster appears, where the electron wave functions are represented as plane waves propagating throughout the crystal [9; 10].

The electrical heterogeneity can be controlled by fluctuations in valence, concentration and temperature [11-13]. For example, the thulium ion reveals a trivalent state in TmS [14; 15], an intermediate valence state in TmSe [16] and a divalent state in TmTe [17]. The electronic configuration of the $Tm^{2+\delta}$ ion is chalcogen dependent. Therefore, the substitution of manganese ion in MnS and MnSe chalcogenides with thulium ions will lead to different electron delocalisation energies. The introduction of non-stoichiometry into the TmSe system favours an increase in the valence of thulium ions to Tm^{3+}.

The aim of the work is to reveal the influence of chalcogen ions on the relaxation time of current carriers, impedance characteristics and their change in a magnetic field.

2. Effect of chalcogen ion on the frequency dependence of impedance

$Tm_xMn_{1-x}S$ solid solutions were synthesised by flux method from polycrystalline manganese sulphide and thulium monosulphide [18]. Samples $(MnSe)_{1-x}(Tm_{0,76}Se)_x$ were obtained by solid-phase reaction method in vacuum quartz ampoules in a single-zone resistivity furnace. The detailed synthesis procedure is described in [19]. The X-ray diffraction study of chalcogenides gives an X-ray diagram corresponding to the NaCl-type FCC lattice.

Impedance, impedance components respond to changes in electronic structure and local lattice deformation, which change the electron density distribution function. Determination of the relaxation time of current carriers is an important characteristic for determining the mechanism of current carrier dissipation. Relaxation can be of both activation and non-activation type [20].

Relaxation of current carriers at frequencies higher than 1 kHz is manifested at temperatures above room temperature, so the frequency dependence of impedance without field and in a magnetic field can be measured at temperatures above room temperature. Figure 1 shows the frequency dependence of impedance $Z(\omega)$ and $Im(Z(\omega))$ for $Tm_{0,05}Mn_{0,95}S$ and $(MnSe)_{1-x}(Tm_{0,76}Se)_x$. When manganese is substituted by thulium ions, the frequencies of the $Im(Z(\omega))$ maxima shift towards the high frequencies in manganese selenide by two orders of magnitude. Regardless of the chalcogen ion, the relaxation time changes dramatically in the vicinity of 400 K (insets in Fig. 1). Below T = 400 K in the system there is one relaxation time in the frequency region 10^2–10^6 Hz and the impedance components are described in the Debye model:

$$\mathrm{Im}\,Z(\omega) = \frac{B\omega\tau}{1+(\omega\tau)^2}, \qquad (1)$$

where τ is the relaxation time of current carriers; B is a parameter. A spectrum of relaxation times appears in the system above 420 K. The relaxation time depends on temperature $\tau = \tau_0 \cdot \exp(E_g/T)$ exponentially with activation energy E_g = 0,47 eV in $(MnSe)_{1-x}(Tm_{0,76}Se)_x$ for x = 0.05.

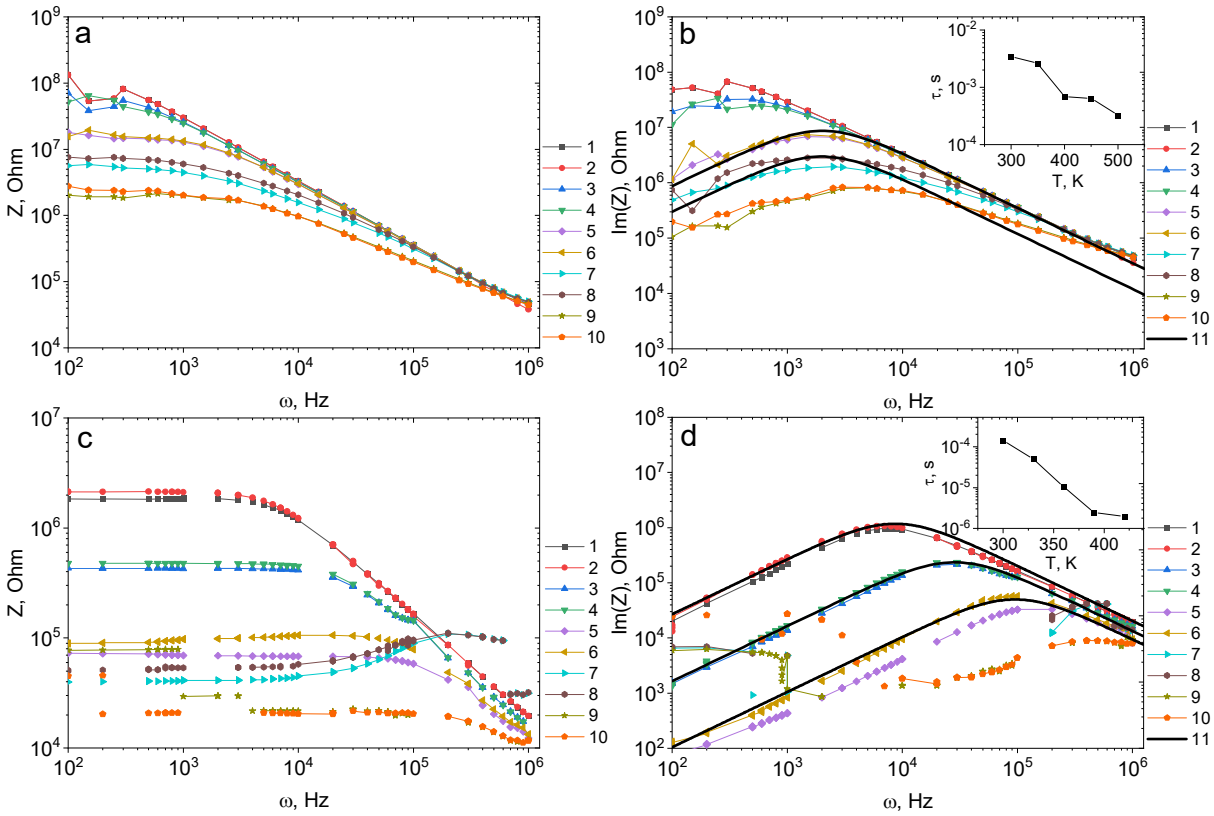

Рис. 1. Частотные зависимости импеданса Z (*a*, *c*) и мнимой части импеданса (*b*, *d*) для образцов $Tm_{0,05}Mn_{0,95}S$ (*a*, *b*), измеренные в нулевом магнитном поле (1, 3, 5, 7, 9) и в поле 8 кЭ (2, 4, 6, 8, 10) при T = 300 (1, 2), 350 (3, 4), 400 (5, 6), 450 (7, 8), 500 K (9, 10), и для образцов $Tm_{0,04}Mn_{0,95}Se$ (*c*, *d*), измеренные в нулевом магнитном поле (1, 3, 5, 7, 9) и в поле 12 кЭ (2, 4, 6, 8, 10) при T = 300 (1, 2), 330 (3, 4), 360 (5, 6), 390 (7, 8), 420 K (9, 10). Вставки: температурные зависимости времени релаксации τ. Результаты эксперимента описаны в рамках модели Дебая (сплошные линии 11)

Fig. 1. Frequency dependences of impedance Z (*a*, *c*) and imaginary part of impedance (*b*, *d*) for $Tm_{0.05}Mn_{0.95}S$ samples (*a*, *b*) measured in zero magnetic field (1, 3, 5, 7, 9) and in a field of 8 kOe (2, 4, 6, 8, 10) at T = 300 (1, 2), 350 (3, 4), 400 (5, 6), 450 (7, 8), 500 K (9, 10) and for $Tm_{0.04}Mn_{0.95}Se$ samples (*c*, *d*) measured in zero magnetic field (1, 3, 5, 7, 9) and in a field of 12 kOe (2, 4, 6, 8, 10) at T = 300 (1, 2), 330 (3, 4), 360 (5, 6), 390 (7, 8), 420 K (9, 10). Inserts: temperature dependences of the relaxation time τ. The experimental results are described within the Debye model (solid lines 11)

The impedance components from frequency in $Tm_{0,1}Mn_{0,9}S$ (Fig. 2, *a*) can be described in the Debye model with a single relaxation time, which has an activation form up to T = 450 K with activation energy E_g = 0.72 eV (insert in Fig. 2, *b*). In $(MnSe)_{1-x}(Tm_{0,76}Se)_x$ with x = 0.1 Im(Z(ω)) is well described in the Debye model:

$$\mathrm{Im}\,Z(\omega) = \frac{A\omega\tau_1}{1+(\omega\tau_1)^2} + \frac{B\omega\tau_2}{1+(\omega\tau_2)^2} \qquad (2)$$

with two relaxation times (Fig. 2, *c*) and with activation energy E_g = 0.6 eV for τ_1 smaller than in $Tm_{0,1}Mn_{0,9}S$.

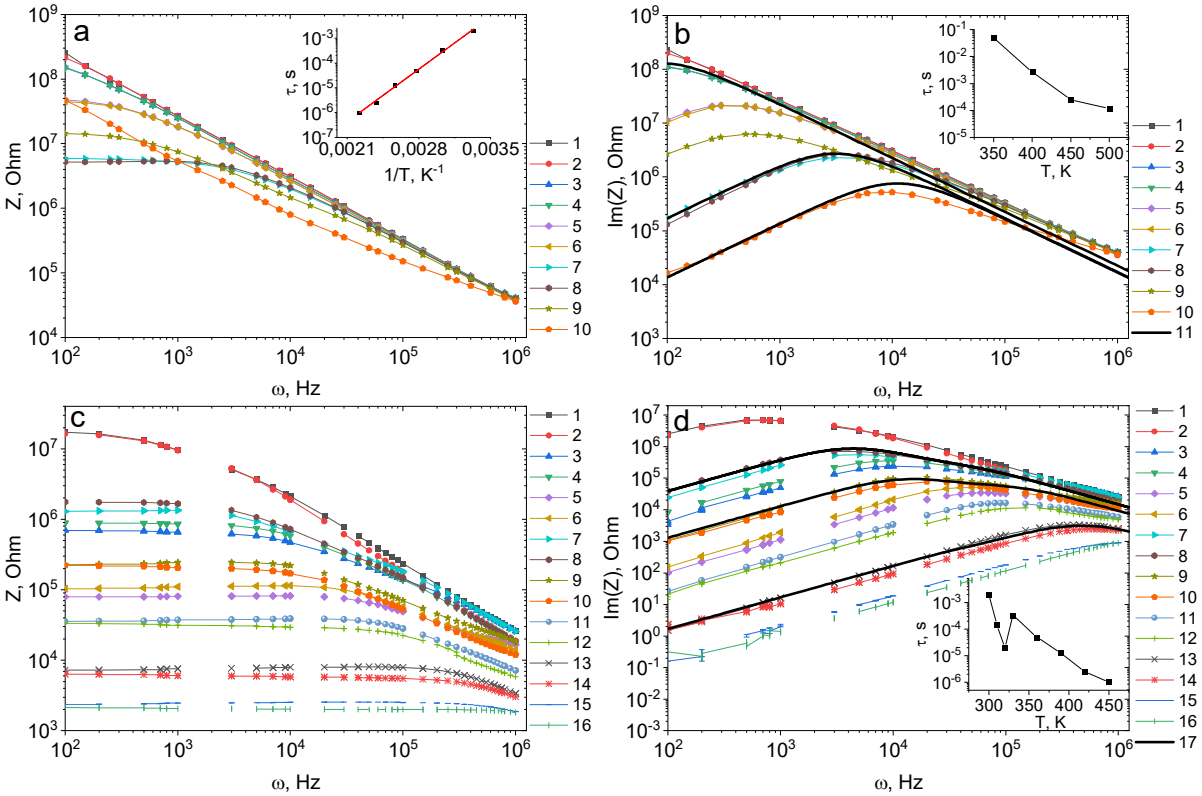

Рис. 2. Частотные зависимости импеданса Z (*a*, *c*) и мнимой части импеданса (*b*, *d*) для образцов $Tm_{0,1}Mn_{0,9}S$ (*a*, *b*), измеренные в нулевом магнитном поле (1, 3, 5, 7, 9) и в поле 8 кЭ (2, 4, 6, 8, 10) при T = 300 (1, 2), 350 (3, 4), 400 (5, 6), 450 (7, 8), 500 К (9, 10), и для образцов $Tm_{0,08}Mn_{0,9}Se$ (*c*, *d*), измеренные в нулевом магнитном поле (1, 3, 5, 7, 9, 11, 13, 15) и в поле 12 кЭ (2, 4, 6, 8, 10, 12, 14, 16) при T = 300 (1, 2), 310 (3, 4), 320 (5, 6), 330 (7, 8), 360 (9, 10), 390 (11, 12), 420 (13, 14), 450 К (15, 16). Вставки: температурные зависимости времени релаксации τ. Результаты эксперимента описаны в рамках модели Дебая (сплошные линии 11, 17)

Fig. 2. Frequency dependences of impedance Z (*a*, *c*) and imaginary part of impedance (*b*, *d*) for $Tm_{0.1}Mn_{0.9}S$ samples (*a*, *b*) measured in zero magnetic field (1, 3, 5, 7, 9) and in a field of 8 kOe (2, 4, 6, 8, 10) at T = 300 (1, 2), 350 (3, 4), 400 (5, 6), 450 (7, 8), 500 K (9, 10) and for $Tm_{0.08}Mn_{0.9}Se$ samples (*c*, *d*) measured in zero magnetic field (1, 3, 5, 7, 9, 11, 13, 15) and in a field of 12 kOe (2, 4, 6, 8, 10, 12, 14, 16) at T = 300 (1, 2), 310 (3, 4), 320 (5, 6), 330 (7, 8), 360 (9, 10), 390 (11, 12), 420 (13, 14), 450 K (15, 16). Inserts: temperature dependences of the relaxation time τ. The experimental results are described within the Debye model (solid lines 11, 17)

3. Magnetoimpedance

The impedance depends on the magnetic field and the chalcogen ion. Fig. 3 shows the changes in impedance in a magnetic field calculated by the formula

$$\Delta Z = (Z(\omega, H) - Z(\omega, H = 0)) / Z(\omega, H = 0)), \quad (3)$$

where Z(ω,H) is the impedance in the magnetic field and without field for Z(ω, H = 0). For substitution concentrations x < 0.05, the impedance increases in the magnetic field and the magnetoimpedance reaches a maximum of ΔZ = 0.35 at T = 450 K for samples $Tm_{0,05}Mn_{0,95}S$ and ΔZ = 0.56 at T = 360 K. With increasing concentration, the change of impedance in the magnetic field decreases. When heated, the magnetoimpedance changes sign in frequency and temperature (Fig. 3, *c, d*).

The impedance in chalcogenides increases in the magnetic field and passes through a maximum when the samples are heated. The increase in impedance is due to the change in the diagonal component of the dielectric permittivity in the magnetic field, which is proportional to the conductivity $\sigma(\omega) = i\omega\varepsilon$, impedance $Z^2 = 1/\sigma^2 + 1 / (\omega C)^2 \approx 1/\varepsilon^2$. In an electrically inhomogeneous medium, the longitudinal component of the dielectric permittivity has the form [21; 22]:

$$\mathrm{Re}[\varepsilon_{xx}(\omega)] = \frac{\varepsilon(1-\beta^2+(\omega\tau)^2(1+\beta^2)^2)}{1+(\omega\tau)^2(1+\beta^2)^2}, \quad (4)$$

where $\beta = \mu H$, μ is mobility, $\tau = RC$. Relative change of impedance [23]

$$\frac{(Z(H)-Z(0))}{Z(H)} = \frac{(\varepsilon(0)-\varepsilon(H))}{\varepsilon(0)} = \frac{\beta^2}{1+(\omega\tau)^2(1+\beta)^2} \quad (5)$$

and its component is satisfactorily described by this function in the region of small concentrations (Fig. 3). As a result, from the impedance it is possible to obtain information about the electrical non-homogeneity [24] and dielectric permittivity in the medium [25; 26].

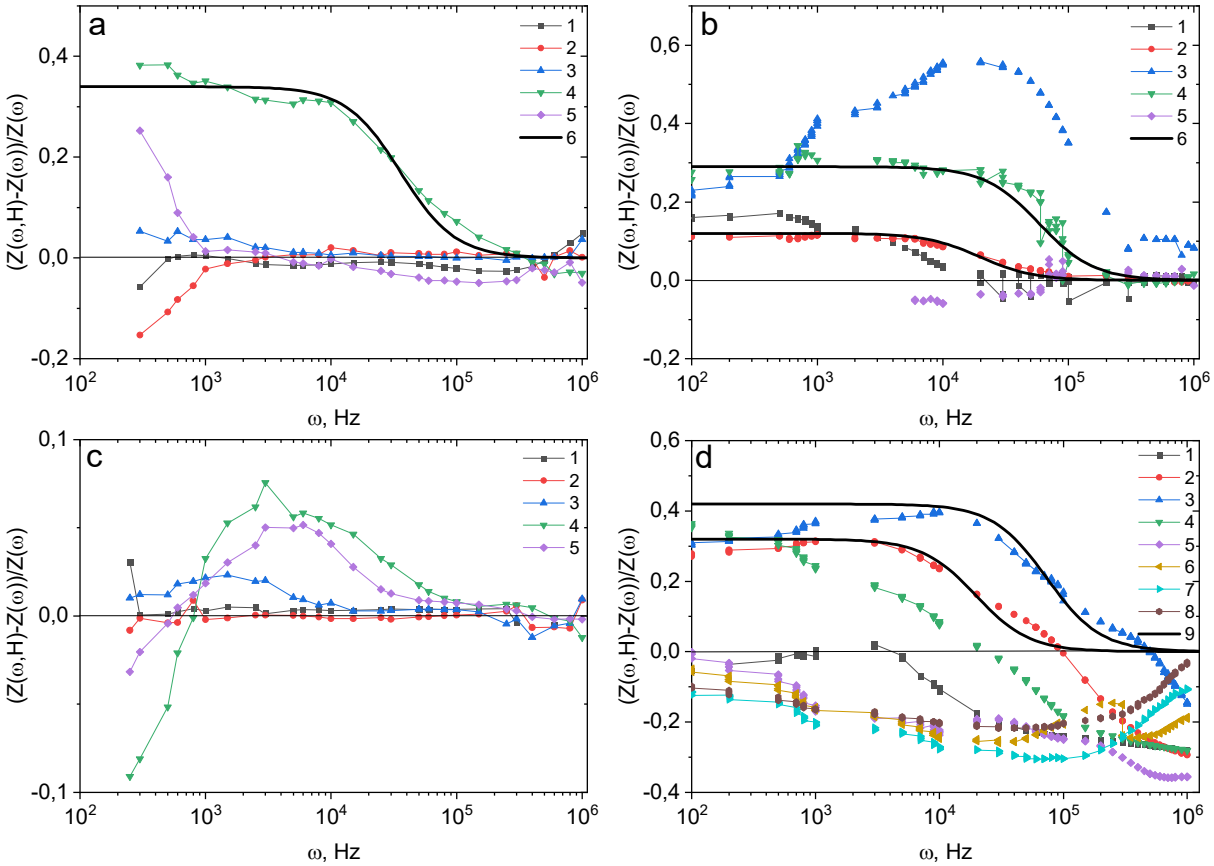

Рис. 3. Магнитоимпеданс в магнитном поле H = 8 кЭ при температурах T = 300 (1), 350 (2), 400 (3), 450 (4), 500K (5) для образцов $Tm_{0,05}Mn_{0,95}S$ (*a*); в магнитном поле H = 12 кЭ при температурах T = 300 (1), 330 (2), 360 (3), 390 (4), 420 K (5) для $Tm_{0,04}Mn_{0,95}Se$ (*b*); в магнитном поле H = 8 кЭ при температурах T = 300 (1), 350 (2), 400 (3), 450 (4), 550 K (5) для образцов $Tm_{0,1}Mn_{0,9}S$ (*c*); в магнитном поле H = 12 кЭ при температурах T = 300 (1), 310 (2), 320 (3), 330 (4), 360 (5), 390 (6), 420 (7), 450 K (8) для $Tm_{0,08}Mn_{0,9}Se$ (*d*). Результаты эксперимента описаны формулой (5) (сплошные линии 6, 9)

Fig. 3. Magnetoimpedance in a magnetic field of H = 8 kOe at temperatures T = 300 (1), 350 (2), 400 (3), 450 (4), 500 K (5) for $Tm_{0.05}Mn_{0.95}S$ samples (*a*); in a magnetic field of H = 12 kOe at temperatures T = 300 (1), 330 (2), 360 (3), 390 (4), 420 K (5) for $Tm_{0.04}Mn_{0.95}Se$ (*b*); in a magnetic field of H = 8 kOe at temperatures T = 300 (1), 350 (2), 400 (3), 450 (4), 550 K (5) for $Tm_{0.1}Mn_{0.9}S$ samples (*c*); in a magnetic field H = 12 kOe at temperatures T = 300 (1), 310 (2), 320 (3), 330 (4), 360 (5), 390 (6), 420 (7), 450 K (8) for $Tm_{0.08}Mn_{0.9}Se$ (*d*). The experimental results are described by formula (5) (solid lines 6, 9)

4. Conclusion

In $(MnSe)_{1-x}(Tm_{0,76}Se)_x$ the prevalence of the reactive part of impedance is found, in the sulfide $Tm_{0,05}Mn_{0,95}S$ the real part of impedance predominates. With increasing concentration of manganese

substitution by tulium, the impedance and its components increase by an order of magnitude. Charge ordering may be formed and the capacitance increases by two orders of magnitude. Relaxation of charge carriers is described in the Debye model. The activation character of relaxation time and activation energy are found. For small concentrations, the impedance increases in magnetic field in chalcogenides. With increasing concentration, the magnetoimpedance changes sign in frequency and temperature. The increase of impedance in the field is caused by the decrease of dielectric permittivity in the magnetic field.

Благодарности

Работа поддержана Российским научным фондом, Правительством Красноярского края и проектом Красноярского научного фонда № 23-22-10016.

Acknowledgements

The study was supported by a grant from the Russian Science Foundation No. 23-22-10016, the Krasnoyarsk Regional Science Foundation.

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Статья поступила в редакцию 07.10.2024; принята к публикации 08.10.2024; опубликована 26.12.2024
The article was submitted 07.10.2024; accepted for publication 08.10.2024; published 26.12.2024