621.315.592

HIGH-FREQUENCY DIELECTRIC MEASUREMENTS ON $TlGa_{1-x}Fe_xSe_2$ SINGLE CRYSTALS

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Frequency dependence of the dissipation factor tanδ, the permittivity ε, and the ac conductivity σ_{ac} across the layers in the frequency range $f = 5\cdot10^4 \div 3.5\cdot10^7$ Hz was studied in layered $TlGa_{1-x}Fe_xSe_2$ single crystals (x = 0; 0.001; 0.005 and 0.01). In the alternate electric fields, the ac–conductivity obeyed the $f^{0.8}$ law at $f = 10^6 \div 10^7$ Hz and the f^n law (where $n = 1.1 \div 2.0$) for $f > 10^7$ Hz. It was established that the mechanism of the ac charge transport across the layers in $TlGa_{1-x}Fe_xSe_2$ single crystals in the frequency range from 10^6 to 10^7 Hz is hopping over localized states near the Fermi level. Estimations yielded the following values of the parameters: the density of states at the Fermi level $N_F = 9.7\cdot10^{17} \div 1.3\cdot10^{18}$ $eV^{-1}\cdot cm^{-3}$; the average time of charge carrier hopping between localized states $\tau = 6.3\cdot10^{-8} \div 3\cdot10^{-7}$ s and average hopping distance R = 190 ÷216 Å.

$TlGaSe_2$ single crystals are typical representatives of layered semiconductors. Layered crystals usually contain structural defects, such as vacancies and dislocations. The presence of these defects results in a high density of localized states near the Fermi level. In [1] it is established by experiments that at T ≤ 200 K in $TlGaSe_2$ along C–axis in constant electric field hopping conductivity with alternating length of jump in localized states near the Fermi level is taken place. Value of state density in vicinity of Fermi level (N_F) calculated from experimental results of $TlGaSe_2$ single crystal dc–conductivity measurement along C–axis is $2\cdot10^{18}$ $eV^{-1}\cdot cm^{-3}$. In [2] it is established that at T ≤ 250 K in $TlGa_{0.99}Fe_{0.01}Se_2$ single crystals along C–axis in dc–electric field a variable range hopping conductivity in forbidden gap near the Fermi level has been taken place and $N_F = 5.6\cdot10^{17}$ $eV^{-1}\cdot cm^{-3}$.

Of some interest is the study of influence of Ga partial substitution in $TlGaSe_2$ for Fe on their dielectric properties in alternate electric fields. This is the aim of the given paper.

Samples of $TlGa_{1-x}Fe_xSe_2$ composition have been synthesized by melting of initial high-purity components in vacuumed quartz ampoules up to 10^{-3} Pa, and their single crystals have been grown by Bridgeman-Stockbarger method. Samples from $TlGa_{1-x}Fe_xSe_2$ for measurements are obtained by spalling along C–axis of the natural spall from massive single crystals and have a thickness $(9.5 \div 12.0)\cdot10^{-3}$ cm. $TlGa_{1-x}Fe_xSe_2$ samples formed flat capacitors whose plane was perpendicular to the crystalline C axis. The capacitor plate area was $(7.3 \div 11.5)\cdot10^{-2}$ cm^2. Ohmic contacts of samples are made by Ag paste.

Measurements of the dielectric coefficients of $TlGa_{1-x}Fe_xSe_2$ single crystals were performed at fixed frequencies in the range $5\cdot10^4 \div 3.5\cdot10^7$ Hz by the resonant method using a TESLA BM 560 Qmeter. For electrical measurements, the samples were placed in a specially constructed screened cell. An ac electric field was applied across the natural layers of $TlGa_{1-x}Fe_xSe_2$ single crystals. The amplitude of the applied field corresponded to the Ohmic region of the current-voltage characteristics of $TlGa_{1-x}Fe_xSe_2$ samples. All measurements were performed at T = 300 K. The accuracy in determining the resonance capacitance and the quality factor Q = 1 / tanδ of the measuring circuit was limited by errors related to the resolution of the device readings. The accuracy of the capacitor graduation was ± 0.1 pF.

The reproducibility of the resonance position was ± 0.2 pF in capacitance and ± (1.0 – 1.5) scale divisions in quality factor.

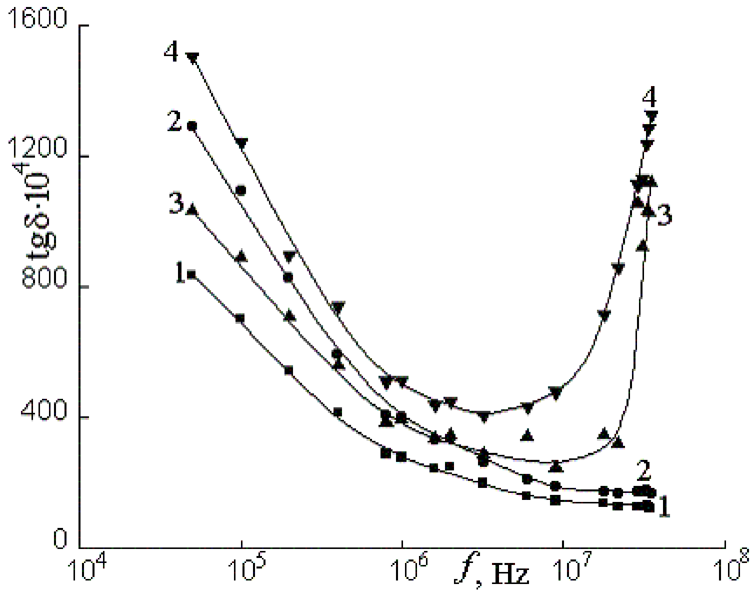

Fig. 1. Frequency dependences of the dissipation factor of $TlGa_{1-x}Fe_xSe_2$ single crystals:

1 – x = 0; 2 – x = 0.001; 3 – x = 0.005; 4 – x = 0.01 (T=300 K).

Fig. 1 shows the experimental frequency dependences of the dissipation factor tanδ for $TlGa_{1-x}Fe_xSe_2$ (x = 0; 0.001; 0.005 and 0.01) single crystals. The tanδ(f) curves have two branches: a steadily descending one and a rising one. A significant dispersion in tanδ is observed for $TlGa_{0.995}Fe_{0.005}Se_2$ and $TlGa_{0.99}Fe_{0.01}Se_2$ (curves 3 and 4). At room temperature, where $TlGa_{1-x}Fe_xSe_2$ single crystals exhibit appreciable ac conductivity, conductivity loss becomes the main dielectric loss mechanism.

We also measured the electric capacitance of $TlGa_{1-x}Fe_xSe_2$ samples in the frequency range $5 \cdot 10^4 \div 3.5 \cdot 10^7$ Hz; the capacitances were 6 ÷ 16 pF. Using the measured capacities of $TlGa_{1-x}Fe_xSe_2$ samples, we calculated the permittivity ε at different frequencies; the permittivity varied from 9 to 17 (Fig. 2).

Fig. 3 shows the experimentally measured frequency dependence of the ac conductivity of $TlGa_{1-x}Fe_xSe_2$ single crystals at T = 300 K. The ac conductivity σ_{ac} varies as

$f^{0.8}$ in the frequency range $3.2 \cdot 10^6 \div 2.9 \cdot 10^7$ Hz for $TlGaSe_2$ and $TlGa_{0.999}Fe_{0.001}Se_2$ single crystals and $8 \cdot 10^5 \div 9 \cdot 10^6$ Hz for $TlGa_{0.995}Fe_{0.005}Se_2$ and $TlGa_{0.99}Fe_{0.01}Se_2$ single crystals. The ac conductivity of investigated crystals at high frequencies ($f > 10^7$ Hz) obeyed the f^n law (where n = 1.1 ÷ 2.0). The $\sigma_{ac} \sim f^{0.8}$ dependence indicates that the mechanism of charge transport is hopping over localized states near the Fermi level [3]. The magnitude of this conductivity is much greater than that of the dc hopping conductivity of studied crystals.

This charge transport mechanism is characterized by the following expression obtained in [4]:

$$\sigma_{ac}(f) = \frac{\pi^3}{96} e^2 kT N_F^2 a^5 f \left[\ln\left(\frac{\nu_{ph}}{f} \right) \right]^4 \qquad (1)$$

where e is the elementary charge, k is the Boltzmann constant, N_F is the density of localized states near the Fermi level, a = 1/α is the localization length, α is the decay parameter of the wave function of a localized charge carrier, $\Psi \sim e^{-\alpha r}$, and ν_{ph} is the phonon frequency. Using expression (1), we can calculate the density of states at the Fermi level from the measured values of the conductivity $\sigma_{ac}(f)$. Calculated values of N_F for investigated

$TlGa_{1-x}Fe_xSe_2$ single crystals are given in Table (localization radius chosen as 34Å, in analogy with the GaSe single crystal [5], which is a double analog of $TlGaSe_2$).

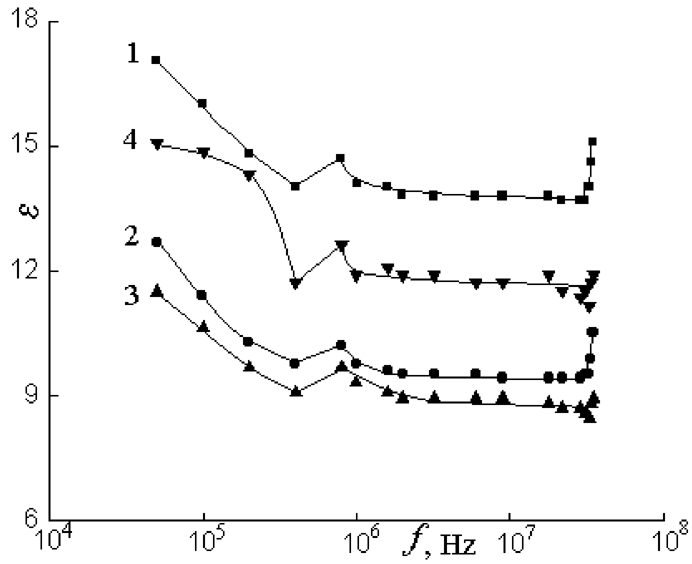

Fig. 2. Frequency dispersion of the permittivity of $TlGaSe_2$ (1); $TlGa_{0.999}Fe_{0.001}Se_2$ (2); $TlGa_{0.995}Fe_{0.005}Se_2$ (3) and $TlGa_{0.99}Fe_{0.01}Se_2$ (4) at T=300 K.

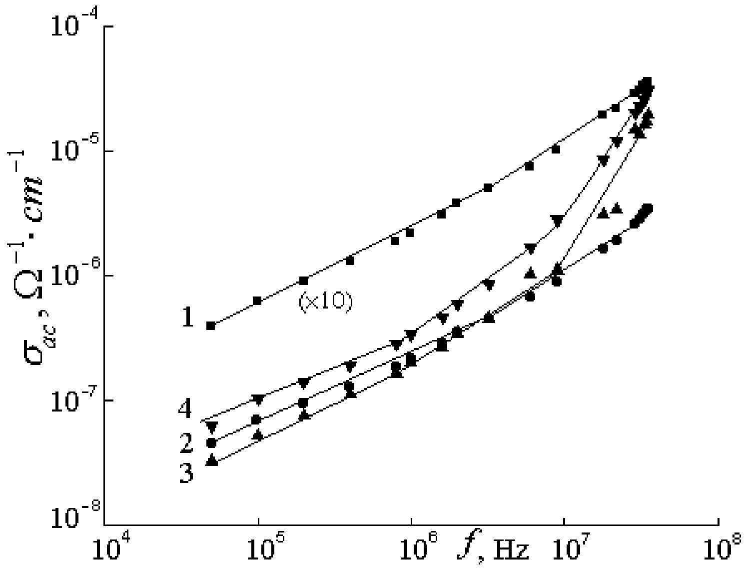

Fig. 3. Frequency-dependent ac conductivities of $TlGa_{1-x}Fe_xSe_2$ single crystals at room temperature.

The values of N_F agrees well with the values N_F found in experiments on the dc conductivity across the $TlGaSe_2$ and $TlGa_{0.99}Fe_{0.01}Se_2$ layers [1, 2].

The theory of ac hopping conductivity provides an opportunity to determine the average time τ of charge carrier hopping from one localized state to another using the formula [3]:

$$\tau^{-1} = \nu_{ph} \exp(-2R\alpha), \quad (2)$$

where R is the average hopping distance:

$$R = \frac{1}{2\alpha} \ln\left(\frac{\nu_{ph}}{f}\right) \quad (3)$$

Calculated values of τ and R are given in Table.

As it is seen from Table, the average hopping distance in $TlGa_{1-x}Fe_xSe_2$ single crystals varied from 190 to 216 Å. The value R calculated from dc conductivity measurements of $TlGa_{0.99}Fe_{0.01}Se_2$ single crystals was equal to 184 Å [2].

As it was shown above at high frequencies $\sigma_{ac} \sim f^2$ in $TlGa_{1-x}Fe_xSe_2$ single crystals. The conductivity proportional to f^2 is related to optical transitions in semiconductors and is dominant at high frequencies [3].

Table. Parameters of $TlGa_{1-x}Fe_xSe_2$ single crystals obtained from high-frequency dielectric measurements

Crystal composition	N_F, $eV^{-1}\cdot cm^{-3}$	R, Å	τ, s	R / a
$TlGaSe_2$	$1.01\cdot10^{18}$	190	$6.3\cdot10^{-8}$	5.6
$TlGa_{0.999}Fe_{0.001}Se_2$	10^{18}	190	$6.3\cdot10^{-8}$	5.6
$TlGa_{0.995}Fe_{0.005}Se_2$	$9.7\cdot10^{17}$	210	$2.0\cdot10^{-7}$	6.18
$TlGa_{0.99}Fe_{0.01}Se_2$	$1.25\cdot10^{18}$	216	$3.0\cdot10^{-7}$	6.35

Thus, the results of high-frequency dielectric measurements on $TlGa_{1-x}Fe_xSe_2$ single crystals are in good agreement with the results of dc conductivity measurements.

1. *Mustafaeva S.N., Aliev V.A., Asadov M.M.* // Fiz. Tverd. Tela (St. Petersburg).1998. N1. P.48-51.
2. *Mustafaeva S.N., Hasanov A.I., Kerimova E.M.* // Izvestiya NAN Azerb. 2003. N5. P. 117-119.
3. *Mott N.F., Davis E.A.* // Electronic processes in Non-Crystalline Materials (Clarendon. Oxford. 1971: Mir. Moscow. 1974).
4. *Pollak M.* // Philos. Mag. 1971. 23. P. 519-542.
5. *Mustafaeva S.N.* // Neorgan. Mater. 1994. N5. P. 619-621.

$TlGa_{1-x}Fe_xSe_2$ MONOKRİSTALLARININ YÜKSƏKTEZLİKLİ DIELEKTRİK ÖLÇMƏLƏRİ

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$TlGa_{1-x}Fe_xSe_2$ (x=0; 0.001; 0.005; 0.01) laylı monokristalların $f=5\cdot10^4\div3.5\cdot10^7$ Hs tezlik oblastında dielektrik itgisinin tangens bucağının dispersiyası (tg δ), dielektrik nüfuzluğu (ε) və monokristalların C oxu istigamətində ac–keziriciliyi (σ_{ac}) tədqiq edilmişdir. Dəyişən elektrik sahəsində 10^6–10^7 Hs tezlik oblastında ac–keçiriciliyi $f^{0.8}$–qanuna tabe olur, $f>10^7$ Hs olduqda isə f^n–qanunu ödənilir ki, harada n=1.1–2.0. Müəyyən olunmuşdur ki, 10^6-dan 10^7 Hs qədər tezliklə $TlGa_{1-x}Fe_xSe_2$ monokristallarında C oxu istigamətində yükün ötürülməsi ilə baş verən kezid mexanizmini yaradan səbəb Fermi səviyyəsinə yaxın olan oblastlarda lokalizə olunmuş yük daşıyıcılarının hoppanmalarıdır. Fermi səviyyəsinə yaxın olan hal üçün, sıxlıg $N_F=9.7\cdot10^{17}\div1.3\cdot10^{18}eV^{-1}\cdot sm^{-3}$, lokalizə olunmuş halların arasındakı hoppanmaların orta vaxtı $\tau=6.3\cdot10^{-8}\div3\cdot10^{-7}$s və hoppanmaların orta məsafəsi R=190÷216 Å təyin edilmişdir.

ВЫСОКОЧАСТОТНЫЕ ДИЭЛЕКТРИЧЕСКИЕ ИЗМЕРЕНИЯ МОНОКРИСТАЛЛОВ $TlGa_{1-x}Fe_xSe_2$

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В слоистых монокристаллах $TlGa_{1-x}Fe_xSe_2$ (x=0; 0.001; 0.005 и 0.01) в частотной области $f=5\cdot10^4\div3.5\cdot10^7$ Гц изучена дисперсия тангенса угла диэлектрических потерь (tg δ), диэлектрической проницаемости (ε) и ac–проводимости (σ_{ac}) поперек слоев. В переменных электрических полях в частотном диапазоне 10^6–10^7 Гц ac–проводимость характеризовалась $f^{0.8}$-законом, а при $f>10^7$ Гц наблюдался f^{n}-закон (где n=1.1–2.0). Установлено, что при частотах от 10^6 до 10^7 Гц за механизм переноса заряда поперек слоев монокристаллов $TlGa_{1-x}Fe_xSe_2$ ответственны прыжки носителей заряда по локализованным вблизи уровня Ферми состояниям. Определены: плотность состояний вблизи уровня Ферми $N_F= 9.7\cdot10^{17}\div1.3\cdot10^{18}$ $эВ^{-1}\cdot см^{-3}$; среднее время прыжков между локализованными состояниями $\tau=6.3\cdot10^{-8}\div3\cdot10^{-7}$ с и среднее расстояние прыжков R=190÷216 Å.