DIELECTRIC PROPERTIES OF Bi₁₂SiO₂₀:Mn CRYSTALS

T.V. PANCHENKO, L.M. KARPOVA, V.M. DUDA

UDC 536.7

Dnipropetrovsk National University, Faculty of Physics (13, Naukova Str., Dnipropetrovsk 49050, Ukraine; e-mail: pancht@mail.ru)

Temperature and frequency dependences of the real and imaginary parts of the complex dielectric permittivity $\varepsilon^*(\omega)$ of Mn-doped Bi₁₂SiO₂₀ crystals in the thermoelectret state have been studied in the audio-frequency range and the temperature interval $300 \div 800$ K. Dielectric anomalies of the quasi-resonance type were observed. The corresponding Cole-Cole diagram demonstrates a section, which is close to linear.

1. Introduction

Photorefractive crystals of the silenite family $Bi_{12}MO_{20}$ (BMO, where M = Si, Ge, or Ti) are successfully applied as active media to various branches of functional electronics. Silenites are characterized by the rather high dielectric permittivity ε ($\varepsilon = 30 \div 160$ at about room temperature [1-3]), the sensitivity of their dielectric properties to the presence of intrinsic defects [3–5] and impurity centers [6–8], and the formation of the electret state [7, 8]. Defects govern the features of the temperature and frequency dependence $\varepsilon(\nu, T)$ and the tangent of the dielectric loss angle, $\tan \delta(\nu, T)$, both in the low ($\nu = 10^2 \div 10^4$ Hz [1, 3, 5-7]) and ultra-high frequency ranges [4]. Some peculiarities are associated in the literature with the processes of relaxation quasi-dipole polarization [5, 7, 8].

Further researches of the dielectric properties of silenites and the possibility of a controlled influence on them by doping seem expedient. We consider the doping of silenites with multivalent ions of irongroup transition metals to be challenging. The latter substitute both M^{4+} and Bi^{3+} ions in the lattice (in oxygen tetrahedra and octahedra, respectively); they are capable to create the centers of strong localization and substantially affect the local level structure in the energy gap and the run of polarization processes.

In this work, we report the results of our study of the dielectric properties of Mn-doped $\rm Bi_{12}MO_{20}$ crystals (BSO:Mn) converted into the thermoelectret state.

2. Experimental Technique

Optically homogeneous BSO:Mn crystals were grown by the Czochralski method. The content of Mn was varied from 0.01 to 0.3 wt.%, the content of uncontrollable impurities did not exceed 10^{-3} wt.%. Specimens were fabricated in the form of $(0.5 \div 1.5) \times 3 \times 7$ -mm³ polished ingots. Pt-electrodes were deposited by cathode sputtering in vacuum onto the (001) faces 3×7 mm² in size. Before experiments, specimens were heated up to a temperature of 800 K and slowly (for about 2 days) cooled down to room temperature in order to establish their electron subsystem into the equilibrium initial state which provided the reproducible results of measurements.

The temperature and frequency dependences of the real, ε' , and imaginary, ε'' , parts of the complex dielectric function ε^* were studied. The measurements of the dependences $\varepsilon'(\nu, T)$ and $\varepsilon''(\nu, T)$ were carried out within the frequency interval $\nu = 10^2 \div 2 \times 10^4$ Hz and the temperature range $T = 300 \div 800$ K with the help of an E8-2 bridge, a GZ-118 generator of sine-wave voltage, and a C1-64A null indicator-oscilloscope. The temperature of the specimens was varied linearly at a rate of 0.16 K/s, making use of a computer-assisted control system. Preliminary polarized and non-polarized specimens were studied. Polarization conditions were varied within the following limits: the voltage U_p = $10 \div 10^3$ V and the temperature $T_p = 300 \div 600$ K; the polarization time of 30 min was constant. Such conditions ensured the formation of a persistent (near room temperature) thermoelectret state [9, 10].

3. Results and Their Discussion

The doping of BSO crystals with Mn ions and the formation of the electret state lead to a specific complicated character of the dependences $\varepsilon'(\nu, T)$ and $\varepsilon'(\nu, T)$ with a lot of features (peaks) that are caused by the contributions of a certain amount of various dielectrically active centers (Fig. 1). In addition, Mn ions induce a 2–5 times reduction of high-temperature losses. The temperature and frequency behavior of the



Fig. 1. Temperature dependences of the real ε' (panels *a* and *c*) and imaginary ε'' (panels *b* and *d*) parts of the complex dielectric permittivity ε^* of BSO:Mn crystals at various frequencies of the measuring field $\nu = 1 \times 10^3$ (panel *a*, curve 1; panel *b*, curves 1 and 1'), 3×10^3 (panel *a*, curve 2; panel *b*, curves 2 and 2'), 8×10^3 (panel *a*, curve 3; panel *b*, curves 3 and 3'), 10^4 (panel *a*, curve 4; panel *b*, curves 2 and 2'), 7×10^2 (panel *c*, curves 1 and 4; panel *d*, curves 1, 1', 4, and 4'), and 5×10^3 Hz (panel *c*, curve 2; panel *d*, curves 2 and 2'). Curves 1', 2', 3', and 4' in panels *b* and *d* are related to the right-hand-side axes. The conditions of preliminary polarization: $U_p = 100$ V (panels *a* and *b*; panel *c*, curves 1-3; panel *d*, curve 1-3; panel *d*, curves 1-3; panel *d*, curve 1-3; panel *d*, curve 1-3; panel *d*, curves 1-3; panel *d*, curve 1-3; panel

parameters ε' , ε'' , and $\tan \delta$ of BSO:Mn also has peculiarities and differs from that known for BSO, BSO:Al, BSO:Ga [6,7], and BSO:Cr [8]. The functions $\varepsilon'(T,\nu)$ and $\varepsilon''(T,\nu)$ are composed of two components each. One of those components hardly depends on the preliminary polarization conditions and forms a monotonous background which determines the crystal (matrix) in whole. Another component consisting of peaks, whose intensities, number, and temperature positions depend on the conditions, under which the electret state of the crystal was formed, is imposed onto this background. In particular, an increase of the preliminary polarization temperature T_p leads to a shift of the temperature spectra $\varepsilon'(T,\nu)$ and $\varepsilon''(T,\nu)$ towards lower temperatures, whereas the general range of their variations remains the same (Fig. 1). In general, the dependences of the dielectric parameters of BMOs on the formation conditions of their thermoor photoelectret states require additional researches. As an attribute of some universality, we note that similar dependences were also observed for BSO crystals, both undoped and doped with Al, Ga, and Cr ions [7,8,11].

3.1. Temperature and frequency behavior of dielectric parameters

The temperature behavior of the real and imaginary parts of the complex dielectric permittivity, $\varepsilon'(T)$ and $\varepsilon''(T)$, was examined in the temperature ranges



Fig. 2. Temperature (a) and frequency (b) dependences of the conductivity of the BSO:Mn crystals converted to the thermoelectret state at $T_p = 300$ (1) and 520 K (2)

 $\Delta T_1 = 300 - 550$ K and $\Delta T_2 = 550 - 750$ K. Within the former, a Π -shaped peak with an abrupt slope on the low-temperature side was observed for the $\varepsilon'(T)$ dependence, as well as a group of peaks with a poorly structured envelope for $\varepsilon'(T)$ and $\varepsilon''(T)$ ones. Intensive peaks in the dependences $\varepsilon'(T)$ manifested themselves in the range ΔT_2 . The dependences $\varepsilon''(T)$ were close to exponential and revealed no intense peaks (Fig. 1), in contrast to BSO:Cr crystals [8]. The temperature behavior of the dielectric parameters of BSO:Mn crystals was inherent to systems with high conductivity, which contributes to $\varepsilon''(T)$, but does not affect $\varepsilon'(T)$. Hence, the high-temperature losses in BSO:Mn occur most likely due to conductivity. This assumption is confirmed by the fact that the crystal electroconductivity calculated making use of the loss factor $\sigma_{\sim} = \varepsilon'' \varepsilon_{\nu} \omega$, where $\varepsilon_{\nu} =$ 8.85×10^{-12} F/m is the vacuum dielectric constant and $\omega = 2\pi\nu$ is the circular frequency, grows exponentially with temperature (Fig. 2, a); and the activation energy of the crystal conductivity $\Delta E_{\sigma} = 0.9$ eV practically coincides with that found for BSO:Mn crystals in dc and ac measurements [12]. We also note the power-law growth of the conductivity as the frequency increases, $\sigma_{\sim} \sim \omega^s$ (s ≈ 0.2 , see Fig. 2,b), which evidences for the hopping mechanism of charge transfer in BSO:Mn crystals [12].

The interesting information concerning the origin of dielectric features (peaks) in the spectra of BSO:Mn crystals is given by the research of their frequency behavior. In both intervals, ΔT_1 and ΔT_2 , the intensities of $\varepsilon'(T,\nu)$ -peaks do not fall down monotonously as the frequency ν grows, but pass through a maximum; the temperature positions of the peaks almost do not shift at that. The intensities of $\varepsilon''(T,\nu)$ -peaks, which, by their positions, correspond to $\varepsilon'(T,\nu)$ -peaks, become redistributed within the ΔT_1 interval, so that they decrease with growth of the frequency. Such a behavior of the $\varepsilon''(T,\nu)$ -peaks would be typical of the peaks of the relaxation origin, but for the absence of their progressive shift towards high temperatures (Fig. 1). Therefore, by their response to the growing frequency, the main features of the dependences $\varepsilon'(T,\nu)$ and $\varepsilon''(T,\nu)$ for BSO:Mn crystals are different from those of the relaxation origin and close to the features of the quasi-resonance type, whereas in BSO, BSO:Al, BSO:Ga [7], and BSO:Cr [8] crystals, on the contrary, they are governed by relaxation processes.

3.2. Cole-Cole diagrams

The complicated character of the $\varepsilon'(T,\nu)$ and $\varepsilon''(T,\nu)$ dependences prohibited us from obtaining the classical diagrams for the $\varepsilon''(\varepsilon')$ dependence in the form of Cole—Cole semicircles at temperatures $T \ge 380$ K. At temperatures $300 \div 370$ K, the monotonous sections of the Cole–Cole diagram $\varepsilon''(\varepsilon')$ were obtained. They are considerably different from the classical ones, inherent to the dispersion of the Debye type, and can be approximated by a straight line $\varepsilon'' = \kappa(\varepsilon' - \varepsilon_{\rm hfl}),$ similarly to what takes place for BSO:Cr crystals [8], but with an essentially larger factor $\kappa = 3.8 \div 4.2$. The magnitude $\varepsilon_{\rm hf1} = 55.8 \ (\varepsilon' \text{ of the matrix})$ was determined from the diagrams $\varepsilon''(\varepsilon')$ by putting $\varepsilon'' \to 0$. We note that the quantity $\varepsilon_{\rm hf1}$ is the high-frequency part of the dielectric permittivity; it is governed by the mechanisms of dispersion that are not engaged in the frequency range concerned.

To analyze the experimental dependences $\varepsilon''(\varepsilon')$ further, we used the coordinates ξ' and ξ'' which are the ε' and ε'' quantities, respectively, normalized by the dielectric contribution: $\xi' = (\varepsilon' - \varepsilon_{\infty}) / (\varepsilon_0 - \varepsilon_{\infty})$ and $\xi'' = \varepsilon'' / (\varepsilon_0 - \varepsilon_{\infty})$, where ε_0 is the static dielectric permittivity, and ε_{∞} is the high-frequency permittivity which has the same sense as the quantity $\varepsilon_{\rm hf1}$ does. Assuming that $\varepsilon_{\infty} = \varepsilon_{\rm hf1} = 55.8$ and $\varepsilon_0 = 70$, we obtained the dependences $\xi''(\xi')$, which were also close to linear (see the inset in Fig. 3). Let us try to find a good approximation in the framework of the classical relaxation dependences.

In the Debye equation $\xi^* = \xi' - i\xi''$, presented by Cole and Cole as a semicircle

$$(\xi' - 0.5)^2 + (\xi'')^2 = 0.25 \tag{1}$$

0,5 on the complex-variable plane (ξ', ξ'') , the variables $= 1/(1 + z^2)$ and $\xi'' = z/(1 + z^2)$, where $z = \omega \tau$ and τ is the relaxation time. 0,4 Deviations from perfect semicircles of type (1) in

various empirical or model equations (Davidson-Cole, Cole–Cole, Havriliak–Nagami, and others) are given by the parameter γ which determines either the interaction between relaxators or their distribution over the relaxation time. One of such equations is the equation for lemniscate-like curves (Fig. 3) which possess sections close to linear in the frequency range that includes the range of our concern [13]:

$$(\xi^*)^{-1} = (1-z)^{\gamma}.$$
 (2)

If $\gamma = 1$, expression (2) converts into the Debye equation. After the separation of the real and imaginary parts of ξ^* , we obtain

$$\xi'(\omega) = \cos(\nu \arctan z) / (1 + z^2)^{0.5\gamma}$$
(3)

and

are ξ'

$$\xi''(\omega) = \sin(\nu \arctan z) / (1+z^2)^{0.5\gamma}.$$
(4)

Taking the experimentally found value for the ratio $\varepsilon^{\prime\prime}/\varepsilon^\prime$ into account, we used the numerical simulation to determine the parameters of the lemniscate-like approximation for the dependence $\xi''(\xi')$. The best parameter set turned out as follows: $\tau = 7.7 \times 10^{-3}$ s and $\gamma = 0.9$ (the inset in Fig. 3). However, the relative error of the approximation by a lemniscate was not less than 20%, whereas it did not exceed 8% for the linear approximation. Thus, the Cole–Cole diagram $\xi''(\xi')$ does have a linear section in the range of sound frequencies.

We note that the magnitude of τ falls within the interval of values, which follow from the Debye relation $\xi''/\xi' = \varepsilon''/(\varepsilon' - \varepsilon_{\rm hf1}) = z$. The assumption that $\varepsilon_{\infty} = \varepsilon_{\rm hf1} = 55.8$ was quite acceptable, because BMO crystals are characterized by large values of ε' at high frequencies, in particular, $\varepsilon' = 43 \div 45$ for undoped BMO at frequencies $\nu = 10^6 \div 10^{11}$ Hz [4,14]. The assumption $\varepsilon_0 = 70$ was based on experimental data.

3.3. Frequency dependences of the dielectric permittivity

The frequency dependences $\varepsilon'(\nu)$ and $\varepsilon''(\nu)$ in the loglog scale have sections that are close to linear and almost parallel to one another (Fig. 4,a). At low frequencies, ε'' grows as ν decreases. Such a behavior, which is not inherent to the classical Debye spectrum, testifies once

ISSN 0503-1265. Ukr. J. Phys. 2006. V. 51, N 11-12



Fig. 3. Lemniscate-like dependence $\xi''(\xi')$ with the parameters $\gamma = 0.9$ and $\tau = 7.7 \times 10^{-3}$ s. The dashed curve denotes the investigated frequency range $\nu = 10^2 \div 2 \times 10^4$ Hz. (Inset) The experimental dependence $\xi''(\xi')$ (insert, 1, dark and light points), linear approximation(insert, 2, solid line), lemniscate-like approximation (insert, 3, dashed curve)

more that the dielectric response includes the contributions from electroconductivity process. The linear and parallel character of the dependences $\lg \varepsilon''$ versus $\lg \nu$ and $\lg \varepsilon'$ versus $\lg \nu$ allows both parts of the complex dielectric function ε^* to be described by a common power-law function of the frequency

$$(\varepsilon'(\nu) - \varepsilon_{hf1}) \sim \varepsilon''(\nu) \sim \nu^{n-1},\tag{5}$$

whose slope angle with respect to the frequency axis determines the value of n. From Eq. (5), one can see that the ratio of two parts of ε^* is a constant which does not depend on the frequency:

$$\varepsilon''(\nu)/(\varepsilon'(\nu) - \varepsilon_{hf1}) = \cot(n\pi/2), \tag{6}$$

where 0 < n < 1 and $\cot(n\pi/2) = \kappa$, i.e. the angle $n\pi/2$ determines the slope of the linear section of the Cole–Cole diagram with respect to the ε' axis. The values $n = 0.15 \div 0.2$ were obtained. A relationship between the dielectric parameters, similar to Eq. (6), is interpreted in the literature as a universal behavior of the dielectric system [15]. It is associated with the hopping mechanism of charge carrier motion over localized states. Such a kind of motion contributes not only to the ac conductivity, but also to the frequency dependence of the dielectric permittivity $\varepsilon'(\nu)$. In this



Fig. 4. Experimental frequency dependences of the real ε' (panel a, curves 1 and 3; panel c, curves 1-3) and imaginary ε'' (panel a, curves 2 and 4) parts of the complex dielectric permittivity of BSO:Mn crystals, and the calculated frequency dependences of the real $\varepsilon'_{\rm osc}$ (panel b, curve 1) and imaginary $\varepsilon''_{\rm osc}$ (panel b, curve 2) parts of the complex dielectric permittivity of the same object at various temperatures T = 290 (panel a, curves 1 and 2), 360 (panel a, curves 3 and 4; panel b, curve 1 and 2), 470 (panel c, curve 1), 600 (panel c, curve 2), and 675 K (panel c, curve 3). Approximation of the dependences ($\varepsilon' - \varepsilon_{\rm hf1}$)(ν) (panel a, curves 1' and 3') and $\varepsilon''(\nu)$ (panel a, curves 2' and 4') by power laws. Curves 3, 3', 4, and 4' are shifted by +1 along the ordinate axis, while curve 2 in panel b by -0.1

case, the peak of dielectric losses must be absent from the $\varepsilon''(\nu)$ spectrum.

Against the background of approximately linear (in log-log coordinates) frequency characteristics, the oscillations of experimental points $\varepsilon'(\nu)$ and $\varepsilon''(\nu)$ were traced (Fig. 4, a). To isolate the contributions of such oscillatory processes, we found the corresponding

dependences $\varepsilon'_{\rm osc}(\nu)$ and $\varepsilon''_{\rm osc}(\nu)$, taking them as the differences between the experimental ($\varepsilon'(\nu) - \varepsilon_{\rm hf1}$ and $\varepsilon''(\nu)$, respectively) dependences and the corresponding calculated dependences of form (5) with n = 0.16($\varepsilon'_{\rm calc}(\nu)$ and $\varepsilon''_{\rm calc}(\nu)$, respectively). The curves $\varepsilon'_{\rm osc}(\nu)$ and $\varepsilon''_{\rm osc}(\nu)$ demonstrate the absence of the significant peaks of dielectric losses up to frequencies of about 5×10^3 Hz and the presence of such a peak in the interval $\nu = 5 \times 10^3 \div 10^4$ Hz (Fig. 4,b). On the other hand, on the removal of the points $\varepsilon'_{\rm osc}$ and $\varepsilon''_{\rm osc}$ (light points in the insert in Fig. 3) from the experimental set, the error of the linear approximation of the dependence $\xi''(\xi')$ would reduce to 3–4% (see the inset in Fig. 3).

Oscillator(quasi-resonance)-type anomalies in the dependence $\varepsilon'(\nu)$ were observed for a number of materials (these were structures with Schottky barriers, based on GaAs crystals, and others) [16, 17]. In the frequency dependences of electrocapacitance, the anomalies revealed themselves as a negative capacitance [17]. They are incompatible with the ordinary relaxation process, where the dependences i(t) of the polarization current on time are monotonous for static fields. However, the nonmonotonic i(t) dependences with sections, where the current falls down or grows up, are compatible with the characteristics of the $\varepsilon'(\nu)$ and $\varepsilon''(\nu)$ dependences of the quasi-resonance type in the frequency range which is determined by the length and the slope of the current-growth sections [17].

It should be noted that the work function amounts to 4.3 eV for BSO crystals [18] and 5.32 eV for Pt electrodes. Therefore, there must emerge a Schottky barrier at the crystal-electrode interface. But the former cannot be responsible for peaks in the $\varepsilon'(T)$ and $\varepsilon'(\nu)$ dependences, because the thermally induced depopulating or filling of the local levels in the energy gap is accompanied solely by the step growth or reduction of the $\varepsilon'(T)$ dependence; the maxima in the dependences $\varepsilon'(\nu)$ are also absent [19].

In undoped BSO crystals, a nonmonotonic variation of the polarization current in time was observed [20], which could stimulate the oscillatory behavior of the $\varepsilon'(\nu)$ dependence. However, the period, for which the current varies, was of about 10^2 s, i.e. oscillations with a frequency of about $10^{-2} \div 10^{-3}$ Hz should be expected. Doping BSO with Mn ions leads to the modification of the energy spectrum of local levels in the energy gap, which can reduce the time interval of the nonmonotonic behavior of the i(t) dependence and make $\varepsilon'(\nu)$ to oscillate at sound frequencies.

The presence of quasi-resonance processes confirms that the frequency dependences $\varepsilon'(\nu)$, which have been measured at those temperatures of the high temperature range $T \ge 450$ K where the curves $\varepsilon'(T)$ peaked, have a maximum (Fig. 4,c). These processes do not contribute to polarization at frequencies of about 10⁴ Hz, and the magnitude of the dielectric permittivity falls down to the value $\varepsilon_{\rm hf2} = 52.7$.

4. Mechanism of Polarization Processes

In the course of polarization processes in BSO:Mn crystals, dielectrically active can be quasi-dipoles of the impurity (i.e. Mn-ion) origin. The research of the optical absorption spectra showed that Mn ions exist in either the Mn^{3+} or Mn^{4+} charged state and occupy mainly the positions of Bi^{3+} ions [21]. In the former case, charge compensation is not necessary, and the difference between ionic radii (1.16 Å for the Bi^{3+} and 0.79 Å for the Mn³⁺ ion) gives rise to a deformation of the surrounding oxygen octahedron and can induce dipole moments in Mn–O complexes. In the latter case, a similar situation becomes complicated owing to the necessity of charge compensation. This compensation can take place, e.g., in the second coordination sphere, where "structure-destroying" $\operatorname{Bi}_{\operatorname{Si}}^{3+}$ ions substitute Si^{4+} ones. Such ions are the well-known intrinsic defects of silenites. The dipole complex made up of Mn_{Bi}^{4+} , O^{2-} , and Bi_{Si}^{3+} ions will be even more complicated. Similar quasi-dipoles in BSO:Cr crystals provoke the existence of peaks in the temperature dependences of internal friction [6] and the classical relaxation behavior of dielectric parameters in the temperature range $T = 400 \div 700$ K [8]. However, the behavior of quasi-dipoles in BSO:Mn crystals does not correspond to the classical mechanism of the thermal relaxation polarization.

At this stage of researches, it seems possible to describe the obtained diagrams $\varepsilon''(\varepsilon')$, which are close to linear, and power-law dependences $\varepsilon''(\nu)$ and $\varepsilon'(\nu)$ in the framework of the universal model proposed by Jonscher for insulators [15]. In this model, the mechanism of polarization is related to the discrete motions of particles in a solid: the jumps of dipoles (quasi-dipoles) between the positions of prevailing orientation or the jumps of charge carriers (electrons and holes). Such a behavior is distinct from that of Debye dipoles and from that given by the band mechanism of conductivity. Another important point in the polarization dynamics is the screening of a dipole (a charge carrier) in a new state of its equilibrium by other dipoles (charge carriers) of the insulating medium. Every jump of a dipole (a charge

DIELECTRIC PROPERTIES OF Bi12SiO20:Mn CRYSTALS

carrier) under the action of an external electric field is coupled with the emergence of a polarization which is reduced afterwards by the following slower screening processes to some extent. The screening efficiency is determined by the parameter p and the power n in the frequency dependences of the real, χ' , and imaginary, χ'' , parts of the dielectric susceptibility χ^* through the relation

$$\frac{\chi''(\omega)}{\chi'(\omega)} = \frac{W_1}{W_2} = \frac{1-p}{p} = \cot\frac{n\pi}{2} = \text{const},\tag{7}$$

where W_1 and W_2 are the energies that were expended and accumulated, respectively, during the period of oscillations; and 0 . If <math>p = 0 (n = 0), the screening is complete and corresponds to the case of the screening by free charges. If p = 1 (n = 1), the screening is absent altogether, which corresponds to the case of immovable charges. The introduction of the parameter p is the simplest manner to take the interaction between charges or dipoles in the insulating medium into account. It follows from general considerations that the screening in dipole systems is weaker than that in systems, where the polarization is induced by charges. Really, the value n = 0.46, which was obtained for BSO:Cr crystals with the predominantly quasi-dipole mechanism of polarization [8], is higher than $n = 0.16 \div 0.2$ for BSO:Mn. Nevertheless, one should bear in mind that the screening efficiency depends also on the concentration of dipoles (charges) and characterizes the force of their interaction with the crystal lattice.

From the Jonscher model, it follows that n corresponds to the exponent power s in the frequency dependence of the ac conductivity:

$$\sigma_{\sim}(\omega) \sim \omega^n. \tag{8}$$

This corresponds to the results obtained (Fig. 2, a), provided s = 0.2. An analogous power law was obtained, while considering the conductivity in percolation theory [22]. It characterizes the mechanism of conductivity as the electron (hole) hopping between localized energy states in the vicinity of the Fermi level. In such a context, the value of n is governed by the spatial and energy distributions of those states, the localization radius of their wave functions, and the hopping multiplicity, i.e. the number of links in the electronhopping chain. The reduction of n is associated with the increase of the hopping multiplicity (e.g., if n = 0.8, jumps occur between the pairs of localized states [22]). Therefore, the magnitude $n = 0.16 \div 0.2$ evidences for a higher-than-double hopping multiplicity in BSO:Mn crystals and testifies that it is larger than that in BSO:Cr crystals [8,23].

All the temperature and frequency dependences of ε' and ε'' (Figs. 1 and 4) testify that the quasi-resonance polarization with the participation of substantially hindered oscillators (quasi-dipoles) makes up a mechanism that is supplementary to the Jonscher one. Low-temperature measurements revealed such oscillators in the frequency range of $(6 \div 10) \times$ 10^3 Hz, their hinderness gives rise to the smearing of the frequency dependences of both the real and the imaginary part of the complex permittivity, as well as to a shift of the maxima of the curves $\varepsilon_{\rm osc}''(\nu)$ and $\varepsilon''(\nu)$ with respect to the curves $\varepsilon'_{\rm osc}(\nu)$ and $(\varepsilon'(\nu) - \varepsilon'_{\rm hfl})$ (Figs. 4, a and b). The transition of BSO:Mn crystals into the thermoelectret state invokes the situation, where quasi-dipoles are located in the internal field of the electret, which reduces their interaction with the nearest environment. This scenario can explain the low-temperature shift of the quasi-resonance peaks in the dependences $\varepsilon'(T)$ and $\varepsilon''(T)$, which accompanies the growth of the temperature of the preliminary polarization T_p , owing to the strengthening of the electret field. Therefore, two mechanisms of polarization - hopping (Jonscher) and quasi-dipole ones - dominate in BSO:Mn crystals at sound frequencies.

- W. Rewald, K. Frick, G.R. Lang, and E. Meier, J. Appl. Phys. 47, 1292 (1976).
- G.M. Safronov, V.N. Batog, Yu.I. Krasilov et al., Izv. AN SSSR, Ser. Neorg. Mater. 6, 284 (1970).
- A.V. Firsov, A.A. Bush, A.A. Maryin et al., Ibid. 21, 446 (1985).
- V.M. Pashkov, Yu.M. Poplavko, and V.M. Skorikov, Fiz. Tverd. Tela 26, 844 (1984).
- J. Link, J. Fontanella, and C.G. Andee, J. Appl. Phys. 51, 4352 (1980).
- M.D. Volnyanskiy, A.Yu. Kudzin, and I.L. Chertkov, Fiz. Tverd. Tela 34, 225 (1992).
- 7. T.V. Panchenko, Ibid. 39, 1223(1997).
- T.V. Panchenko, L.M. Karpova, and V.M. Duda, Ibid. 42, 671 (2000).

- T.V. Panchenko and L.M. Karpova, Izv. RAN, Ser. Fiz. 67, 1153 (2003).
- T.V. Panchenko and L.M. Karpova, Fiz. Tverd. Tela 40, 472 (1998).
- T.V. Panchenko and G.V. Shezhnoy, Ferroelectrics 174, 51 (1995).
- V.T. Avramenko, A.Yu. Kudzin, T.V. Panchenko, and G.Kh. Sokolyanskiy, Poluprov. Elektr. 3, 139 (1984).
- Yu.M. Poplavko, *Physics of Insulators* (Vyshcha Shkola, Kyiv, 1982) (in Russian).
- Y.R. Reddy and L. Sirdeshnrukh, Phys. Stat. Solidi A 103, K157 (1987).
- A.R. Jonscher, Sci. Papers of the Inst. of Electrical Engineering Fundamentals, Ser. Współpraca 16, 5 (1977).
- 16. B.S. Doyle, J. Phys. D 19, 1129 (1986).
- 17. A.K. Joncher, J. Chem. Soc., Faraday Trans. 2 82, 75 (1986).
- V.I. Chmyrev, I.V. Tsisar, V.M. Skorikov, and A.Ya. Vasilyev, Neorg. Mater. 29, 262 (1993).
- L.S. Berman and A.A. Lebedev, Capacitance Spectroscopy of Deep Centers in Semiconductors (Nauka, Leningrad, 1981) [in Russian].
- 20. A.E. Attard, J. Appl. Phys. 69, 44 (1991).
- T.V. Panchenko and S.Yu. Kopylova, Ferroelectrics 322, 69 (2005).
- N.F. Mott and E.A. Davies, *Electronic Processes in Non-Crystalline Materials* (Oxford University Press, Oxford, UK, 1979).
- V.I. Kalinin, Zh.S. Kuchuk, N.G. Gorashchenko, and A.A. Mayer, Izv. AN SSSR, Ser. Neorg. Mater. 24, 637 (1988).

Received 27.12.04, revised version - 10.08.06. Translated from Ukrainian by O.I. Voitenko

ДІЕЛЕКТРИЧНІ ВЛАСТИВОСТІ КРИСТАЛІВ Ві12SiO20:Мn

Т.В. Панченко, Л.М. Карпова, В.М. Дуда

Резюме

У діапазоні звукових частот і в області температур 300—800 К досліджено температурно-частотні характеристики дійсної та уявної частин комплексної діелектричної проникності $\varepsilon^*(\omega)$ кристалів Bi₁₂SiO₂₀:Мп у термоелектретному стані. Спостережено діелектричні особливості квазірезонансного типу, одержано близьку до лінійної ділянку в діаграмі Коул—Коула.