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RESEARCH OF THE ELASTIC WAVE VELOCITY DISPERSION IN X-RAY-IRRADIATED LiF CRYSTALS

The influence of a preliminary deformation (the residual strain $\varepsilon = 0.65\%$) and x-ray irradiation to exposure doses of 0–800 R on the frequency dependence of the sound velocity, $v(f)$, in LiF crystals in the frequency interval from 7.5 to 232.5 MHz and at room temperature has been studied using the pulsed technique. By extrapolating the results obtained for $v(f)$ to the low-frequency interval and using the well-known theoretical relations, the coefficient of dynamic viscosity B and the dislocation density Λ were found to be independent of the irradiation dose. At the same time, the absolute values of B were found to be lower and the values of Λ higher by an order of magnitude than the corresponding values obtained with the use of the most reliable techniques, such as the methods of high-frequency internal friction and etch pit counting, respectively.

Keywords: irradiation, dislocation density, straining, Burgers vector, shear modulus, average effective length of a dislocation segment.

1. Introduction

Experimental researches of the processes that give rise to the fixation of highly mobile dislocations by radiation-induced defects and the elucidation of their influence on the frequency dependence of the ultrasound velocity, $v(f)$, are rather important. On the one hand, they allow one to obtain the data on the elastic properties of crystals, which reflect the character of the interparticle coupling [1]; on the other hand, they provide an opportunity to study the nature of the interaction between dislocations and elementary excitations in the crystal [2]. It should be noted that the experimental dependences $v(f)$ were earlier studied only for NaCl [3, 4] and LiF [5] crystals. The acoustic method used in works [3–5] turned out rather informative. According to the dislocation theory [6], this method can be used to determine the key parameters of a dislocation structure, such as the dislocation density Λ and the average effective length of a dislocation segment L, provided that the constant of dynamic dislocation damping B is known.

From the acoustic measurements carried out in the frequency interval 10–100 MHz for NaCl crystals with the residual strain $\varepsilon = 0.06\%$, the authors of work [3] found that the deformation increases the dispersion from 0.5 to 4% and shifts the dispersion interval toward lower frequencies. While treating their results in the framework of theory [6] and using the adopted value $B = 1.2 \times 10^{-5}$ Pa × s, the authors of work [3] obtained the value $\Lambda = 3 \times 10^{11}$ m^{-2}, which turned out substantially overestimated in comparison with the value Λ_e given for the dislocation density by the method of pit etching in crystals strained to 0.06%.

To make those results more accurate, the dependence $v(f)$ was carefully measured in work [4], again for NaCl crystals but in a much wider frequency interval from 7.5 to 217.5 MHz, when the residual deformation was varied in the interval from 0.2 to 1%, and under x-ray irradiation to a dose of 300 R. As a result of researches, the effect of inverse shift for the dispersion curves $v(f)$ was revealed for the first time, which consists in that the frequency curves, when the residual deformation of the specimen increases, first shift toward low frequencies and, after the x-ray irradiation of the specimen, start to move in the opposite direction. In addition, having determined the value of Λ_e by counting the etch pits, the author of work [4] reliably found that the constant of dislocation damping B does not depend on the dislocation density. At the same time, it was marked that the absolute value of B is considerably underestimated in comparison with the value of B_e determined in work [7] using the conventional "reference" technique of high-frequency internal friction, i.e. from the descending branch in the frequency dependence of the dislocation decrement $\Delta_d(f)$ for the crystals of the same batch.

In the recent work [5], our aim was to verify the presence of effects observed in works [3, 4] and the validity of the results obtained by the cited authors for other crystals, in particular, LiF. For this purpose, the frequency dependences of ultrasound velocity in crystals were studied using non-strained specimens, specimens strained to $\varepsilon = 0.65\%$, and strained specimens irradiated to 132 R. In accordance with the results of works [3, 4], the deformation gave rise to a shift of the $v(f)$ curve toward low frequencies, whereas irradiation, as was observed in work [4], resulted in the inverse shift of this curve. In addition, in spite of the preliminary character of experiment [5], LiF specimens, similarly to NaCl ones [4], demonstrate a tendency for the parameters B and Λ to be independent of the irradiation dose. In work [5], a reduction of the average effective length of a dislocation segment L from 13.5×10^{-7} to 10.6×10^{-7} m under the action of irradiation was also registered. This fact is in good qualitative agreement with the results of work [8], where the specimens of the same batch were used to study the structural characteristics of crystals (from the high-frequency asymptote of the above-mentioned resonance curve $\Delta_d(f)$).

Taking all that into account, we continue, in this work, the researches started in work [5] devoted to the influence of x-ray irradiation on the dispersion dependences $v(f)$ in LiF crystals. Those dependences can be used not only to calculate the dynamic, B, and structural, Λ and L, dislocation characteristics, but also to determine the limits of theory [6] applicability, when the results obtained for $v(f)$ are extrapolated to the low-frequency region.

2. Experimental Technique

Here, we studied the influence of low doses of long-wave x-ray radiation on the frequency dependence of the elastic wave velocity, $v(f)$, in the frequency interval of 7.5–232.5 MHz in LiF crystals with the residual strain $\varepsilon = 0.65\%$ and at $T = 300$ K. The propagation velocity of ultrasound waves was measured in the pulse regime, by using the selector method on an installation described in work [9]. In experiments, we used specimens with a purity of 10^{-4} wt.%, with the crystallographic orientation $\langle 100 \rangle$, and $17 \times 17 \times 29$ mm^3 in dimension.

According to the technology described in works [4, 5, 8], the specimens to study, after their cut-off, were finely polished to achieve the nonparallelism of working surfaces of approximately 1 μm/cm, which was monitored with the help of an IKV optimeter. The surface nonparallelism in the system "piezoquartz–sticker–specimen" could also be estimated independently when imposing the exponential reference signal on a series of reflected pulses observed on the oscilloscope screen in the course of crystal sounding. To remove the internal stresses that could emerge owing to a mechanical treatment of the specimens, the latter were annealed in a muffle furnace MP-2UM for 12 h at a temperature of about $0.8T_{melt}$ and, then, slowly cooled down to room temperature. For highly mobile dislocations to be introduced into the crystal, the latter was preliminarily deformed to achieve the residual strain $\varepsilon = 0.65\%$. At the indicated values of experimental parameters, the dispersion curve $v(f)$ had such initial frequency position, from which it was convenient to observe its further shift toward high frequencies in the course of dislocation fixation by radiation-induced defects.

The achievement of the required residual deformation was checked up by means of the exact registration of the crystal yield point on a tape recorder KSP-4. The working length of the crystal before and after deformation was monitored to an accuracy of 1 μm with the help of a comparator IZA-2. The specimens were deformed by squeezing them on an Instron machine at a rate of about 10^{-5} s^{-1}. In this regime of deformation [7], no slip bands arise, and the etch pits regularly cover the crystal surface, which enables the dislocation density Λ_e to be accurately determined with the use of the software Photoshop.

The procedure of specimen irradiation with x-rays did not differ from that described in works [5, 10].

3. Experimental Results and Their Discussion

In Fig. 1, the experimental dependences $v(f)$ are depicted which were measured for LiF crystals: non-strained (curve *1*), preliminarily strained to $\varepsilon = 0.65\%$ (curve *2*), and strained and irradiated afterward with x-rays for 20–120 min (curves *3* to *6*). One can see that the character of the frequency dependences $v(f)$ substantially changes, when the dislocation structure of the crystal transits from one state into another one. At first, when the "growth" dislocations were fixed by impurity atoms in the non-deformed (annealed) crystal, the propagation veloc-

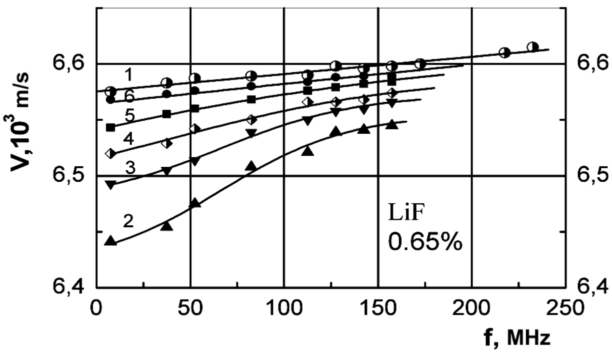

Fig. 1. Frequency dependences of the ultrasound propagation velocity in LiF crystals for (*1*) a non-strained specimen, (*2*) a specimen with a residual deformation of 0.65%, and (*3* to *6*) a specimen deformed to 0.65% and x-ray-irradiated for 20, 40, 60, and 120 min, respectively

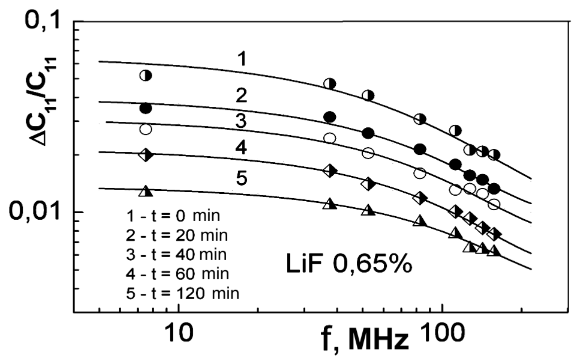

Fig. 2. Frequency dependences of the modulus defect in LiF crystals (*1*) deformed to 0.65%, and (*2* to *5*) x-ray-irradiated for 20, 40, 60, and 120 min, respectively. Solid curves are the theoretical curves taken from work [11]

ity of acoustic waves changed practically linearly as the frequency grew (curve *1*). Then, with the appearance of a significant number of highly mobile dislocations in the crystal owing to its deformation to 0.65%, the character of the curve $v(f)$ drastically changed (curve *2*). Namely, the frequency dependence $v(f)$ acquired a sigmoidal profile, and there emerged a strongly pronounced interval with dispersion, which is especially notable in the low-frequency section.

If the same specimen is further irradiated to various exposure doses within the interval of approximately 130–800 R, the frequency dependences $v(f)$ (curves *3* to *6*) become considerably shifted toward higher frequencies, approaching closer and closer the frequency position of curve *1* for the non-deformed specimen. The results presented in Fig. 1 can be interpreted in accordance with work [6] as follows. At low frequencies, the dislocation moves in phase with the applied stress, and the actual stiffness of the crystals turns out lower than that of dislocation-free ones. As the frequency grows, the synchronism between the motions of a dislocation and the exciting external field becomes substantially broken, and the elastic modulus reaches its true value.

After the set of dispersion curves $v(f)$ had been obtained, we plotted the corresponding dependences of the modulus defect, $\Delta C_{11}/C_{11}$, on the frequency f (see Fig. 2). The figure demonstrates that the experimental curve *1* plotted for the crystal deformed to 0.65% has the maximum amplitude of the modulus defect $\Delta C_{11}/C_{11}$, which smoothly decreases, as the frequency grows. However, if the same specimen was irradiated for 20–120 min, the experimental curves (*2* to *5*) decrease by amplitude and are shifted toward higher frequencies.

Unlike Fig. 1, the array of experimental points in Fig. 2 was approximated by the frequency dependence of the modulus defect calculated in work [11] for the function $(\Delta C_{11}/C_{11})(f)$ in the approximation, when the dislocation segments are distributed exponentially over their lengths. One can see that the experimental and theoretical data are in satisfactory agreement with one another. Imposing the theoretical profile on the experimental data made it possible to determine its frequency and amplitude positions, which allowed us to determine, in a well-founded manner and directly from the theoretical curve, the values of the frequency, f_0, and the modulus defect, $(\Delta C_{11}/C_{11})_0$, corresponding to the points of the curves in Fig. 2, where their linear behavior terminated. These are those "reference" points, where the modulus defect $(\Delta C_{11}/C_{11})_0$ extrapolated to the low-frequency region starts to acquire its maximum value. This procedure used for the determination of the parameters f_0 and $(\Delta C_{11}/C_{11})_0$ allowed us to plot Fig. 3.

We note that the modulus defect was calculated in this work using the relation $2\Delta V/V_\infty = \Delta C_{11}/C_{11}$, where $\Delta V = V_\infty - V$. The purely elastic velocity $V_\infty = 6.61 \times 10^3$ m/s was measured at a frequency of 217.5 MHz, when the crystal behaves itself as an ideal one owing to the absence of dislocation effects. From Fig. 3, one can see that the increase of the irradiation exposure dose gives rise, on the one hand, to a reduction of the modulus defect

$(\Delta C_{11}/C_{11})_0$ and, on the other hand, to the growth of the frequency f_0. It should be noted that the absolute values of ultrasound velocity in the frequency interval from 7.5 to 232.5 MHz were measured to an accuracy of 0.05–0.1% [9], whereas the modulus defect $(\Delta C_{11}/C_{11})_0$ and the frequency were determined from the data (Fig. 2) with an accuracy of 5–7 or 15–20%, respectively.

On the basis of experimental results depicted in Fig. 3 and using the theoretical relations describing the low-frequency branch of the dislocation resonance [6], it is possible to calculate the coefficient of dislocation damping B and to determine the main parameters of the crystal dislocation structure, Λ and L. According to the theory [6], the formula for the modulus defect extrapolated to the low-frequency region looks like

$$\Delta C_{11}/C_{11} = (6\Omega\Delta_0\Lambda L^2)/\pi. \tag{1}$$

Substituting the expressions $\Delta_0 = 8Gb^2/(\pi^3 C)$ and $L^2 = (0.084\pi C)/(2Bf_0)$ taken from work [6] into Eq. (1), we obtain a relation for the calculation of Λ in the form

$$\Lambda = \frac{\pi^3 f_0 B_e}{2.016\Omega G b^2}(\frac{\Delta C_{11}}{C_{11}})_0, \tag{2}$$

where Ω is the orientation factor, G the shear modulus, b the magnitude of Burgers vector, $(\Delta C_{11}/C_{11})_0$ the value of modulus defect measured at the frequency f_0, and B_e the constant of dislocation damping.

Carrying out the calculations by formula (2), in which we take $\Omega = 0.311$ and $Gb^2 = 28.77 \times 10^{-10}$ Pa $\times$ m^2 [5,8], the experimental data exhibited in Fig. 3, and the damping constant $B_e = 3.7 \times 10^{-5}$ obtained recently for the researched crystals in work [8], we obtain the dependence $\Lambda(t)$ (Fig. 4, curve *1*). The invariance of the dislocation density Λ observed experimentally at various exposure doses of irradiation is quite expectable, because the doses applied in our experiments were negligibly low in comparison with those that could stimulate a crystal deformation [12]. At the same time, the average value $\Lambda = 1.82 \times 10^{11}$ m^{-2} calculated by formula (2) raises some doubts, because it is by an order of magnitude larger that the corresponding value $\Lambda_e = 1.74 \times 10^{10}$ m^{-2} found directly by counting the etch pits on the crystal surface [13].

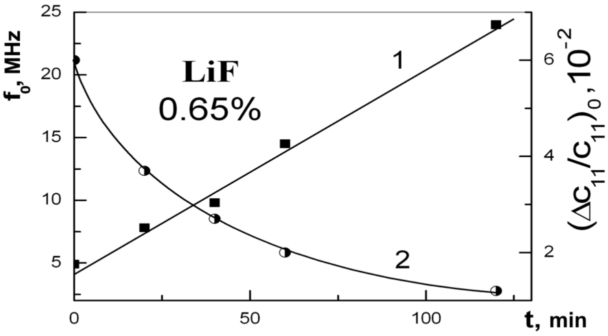

Fig. 3. Dependences of (*1*) the frequency f_0 and (*2*) the modulus defect $(\Delta C_{11}/C_{11})_0$ on the irradiation time of LiF crystals

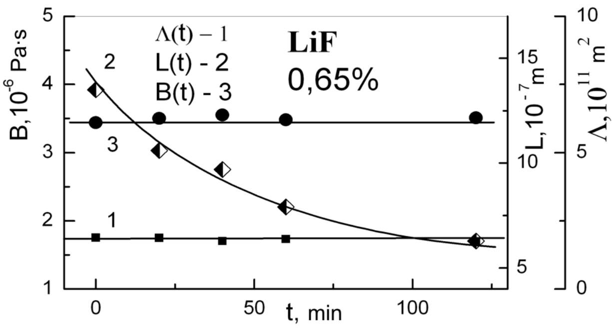

Fig. 4. Dependences of (1) the dislocation density Λ, (*2*) the average effective length of dislocation segments L, and (*3*) the coefficient of dislocation damping B on the irradiation time of LiF crystals

After the base of experimental data had been created, the calculation of another parameter of the dislocation structure, L, in the framework of the theory [6] by the formula

$$L = \sqrt{\frac{0.084Gb^2}{B_e f_0(1-\nu)}} \tag{3}$$

became possible. Here, ν is Poisson's ratio. Substituting $\nu = 0.27$ [5, 8] into formula (3), we calculated the functional dependence $L(t)$ shown in Fig. 4 by curve *2*. As was expected, a gradual increase of the exposure dose gives rise to a monotonic decrease of the effective length of a dislocation segment, L, owing to its fixation by radiation-induced defects. However, the initial value of L obtained before the irradiation was approximately 1.5 times larger than the value determined in work [8] on the basis of equations describing the position of the dislocation resonance.

By solving Eq. (2), we obtain the following expression for the coefficient of dislocation damping:

$$B = (2.016\,\Omega G b^2 \Lambda_e)/(f_0\,(\Delta C_{11}/C_{11})_0\,\pi^3). \quad (4)$$

The results of calculations by this formula show that the damping coefficient B for LiF crystals, similarly to what was obtained in work [8], does not depend on the irradiation time, which is illustrated by curve *3* in Fig. 4. However, the absolute value of the constant B turned out by an order of magnitude smaller than the value of B_e determined using the method of high-frequency internal friction [8] by analyzing the descending branch of the dislocation resonance. The determined dependence $B(t)$ agrees with the theoretical conclusions [2] that the dynamic dislocation damping in LiF at a constant temperature is governed only by dissipative processes in the phonon subsystem of the crystal.

From the analysis of experimental data, it follows that the obtained results concerning the revealed independence of the quantities B and Λ of the irradiation time t and a decrease of the dislocation mobility, which manifests itself in a reduction of L with the growth of the irradiation dose, qualitatively coincide with the data obtained, by using the well-known methods of researches mentioned above. However, there exists a considerable quantitative discrepancy between the values determined in this work and those obtained in work [8]; this is especially true for the absolute estimates of the quantities B and Λ. A preliminary comparative analysis of indicated experimental data allows us to suppose that the most plausible origin of the substantial difference between the calculated B and Λ values and the corresponding B_e and Λ_e ones obtained in the framework of conventional techniques consists in that the relation $\omega/\omega_m \ll 1$, where ω and ω_m are frequencies corresponding to the low-frequency branch of the dislocation resonance $\Delta_d(f)$ and its maximum, respectively, is not obeyed under the given experimental conditions. The condition $\omega/\omega_m \ll 1$ is known to form the basis of simplifications carried out while deriving formulas (1)–(3) in the framework of the dislocation theory [6]. On the other hand, those data confirm the conclusion made by the authors of work [14] that it is impossible to describe experimental data for the indicated frequency ranges in the framework of the theory [6], by engaging only a unique mechanism of ultrasound absorption. In accordance with work [14], the results of measurements can be described by a general frequency profile only if different B values are used for every frequency branch. In order to determine the applicability limits for the theory [6] in more details, the results of high-frequency measurements should be appended with the data on the absorption coefficient and the modulus defect measured at low frequencies.

4. Conclusions

The influence of long-wave x-ray irradiation in low doses on the ultrasound velocity dispersion, $v(f)$, in LiF crystals with a residual strain of 0.65% was studied in the frequency interval from 7.5 to 232.5 MHz and at $T = 300$ K. The preliminary deformation of specimens was found to result in the appearance of a pronounced dispersion region in the curve $v(f)$, which had a linear character for non-deformed specimens. The dispersion phenomenon was observed to be especially pronounced at low frequencies, when the scattering of the elastic energy of ultrasound waves is governed by a high mobility of long dislocation loops. Irradiation of those crystals to exposure doses of 0–800 R was found to result in a gradual reduction of the dispersion region and its shift toward higher frequencies, which can be explained by the pinning of dislocations by radiation-induced defects.

On the basis of the results obtained for LiF for the first time and using the method of extrapolation of the results on $v(f)$ to the low-frequency interval, it is established that the constant of dislocation damping B and the dislocation density Λ remain constant as the irradiation time t grows. The obtained dependence $B(t)$ confirms our theoretical prediction that the dynamic damping of dislocations at a constant temperature is governed only by dissipative processes in the phonon subsystem of the crystal.

The analysis of the results obtained show that the absolute value of B is lower and the value of Λ is larger by an order of magnitude in comparison with the corresponding values B_e and Λ_e obtained in the framework of the conventional methods of high-frequency internal friction and selective etching of the crystal surface, respectively. The preliminary common analysis of the indicated experimental data allows us to suppose that their mismatch originates from the invalidity of the condition $\omega/\omega_m \ll 1$, where ω and ω_m are the frequencies corresponding to the low-frequency branch of the dislocation resonance $\Delta_d(f)$ and its maximum, respectively. Just this primary condition was used to make simplification in the dislocation theory mentioned above, while deriving formulas (1)–(3).

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ДОСЛІДЖЕННЯ ДИСПЕРСІЇ
ШВИДКОСТІ ПРУЖНИХ ХВИЛЬ
В ОПРОМІНЕНИХ КРИСТАЛАХ LiF

Р е з ю м е

Імпульсним методом в області частот 7,5–232,5 МГц при T = 300 К досліджено вплив попередньої деформації (ε = = 0,65%) і опромінення (в інтервалі доз 0–800 Р) на хід частотних залежностей швидкості ультразвуку в кристалах LiF. Екстраполюючи результати по дисперсії швидкості v (f) на область низьких частот та використовуючи відомі теоретичні співвідношення було встановлено, що коефіцієнт динамічної в'язкості B і густина дислокацій Λ зі зростанням дози опромінення залишаються незмінними. Разом з тим виявлено, що абсолютне значення величини B є в 10 разів меншим, а величини Λ – у стільки ж разів більшим від тих значень, що дають найбільш коректні методи – високочастотного внутрішнього тертя і прямого підрахунку ямок протравлювання відповідно.