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THE INFLUENCE OF X-RAY IRRADIATION ON ELASTIC, DYNAMICAL AND STRUCTURAL CHARACTERISTICS OF STRAINED LiF CRYSTALS

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The influence of a preliminary deformation $\varepsilon = 0.4\%$ and X-ray irradiation to exposure doses of 400 and 800 R on the frequency dependence of the sound velocity *v(f)* in LiF crystals in the frequency interval 37.5…172.5 MHz and at room temperature has been studied using the pulsed technique. The coefficient of dynamic viscosity *B* and the dislocation density Λ were found to be independent of the irradiation dose. 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.

INTRODUCTION

It is greatly important to investigate the processes that lead to the fixation of high-level dislocations with the help of detected defects caused by radiation and the determination of their influence on the frequency dependence of the ultrasonic velocity, *v(f)*. First, they give us the opportunity to obtain data on the elastic properties of crystals reflecting the nature of the interparticle bonds [1]; and secondly, they open the possibilities of studying the nature of the interaction between dislocations and elementary excitations in the crystal [2]. We can't help saying that the experimental dependences *v(f)* were earlier analyzed only for NaCl [3, 4] and LiF [5, 6] crystals. The acoustic method used in works [3–6] turned out rather informative. According to the dislocation theory [7], 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. The effect of inverse shift for the dispersion curves *v(f)* was revealed for the first time, and it is as follows the frequency curves, when the residual deformation of the specimen increases, first shift aligned towards low frequencies and, after the X-ray irradiation of the specimen, it starts to move in the opposite direction. In addition, having determined the value of Λ_e by counting the etch pits, the author of work [4–6] reasonably proves 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 [8] 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 works [5, 6], our main goals were the verifying of the presence of effects observed in work [4] and the fitting of the results obtained by the cited authors for other crystals, in particular, LiF. For this purpose, the frequency dependences of ultrasonic velocity in crystals were investigated using non-strained specimens, specimens strained to $\varepsilon = 0.65\%$, and strained specimens irradiated to 800 R. Along 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. Moreover 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, 6], a reduction of the average effective length of a dislocation segment *L* under the action of irradiation was also registered. This fact is in good qualitative agreement with the results of works [8, 9], 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 $\Delta_d(f)$). Based on all of the foregoing, we continue, in this work, the researches started in work [5, 6] devoted to the influence of X-ray irradiation on the dispersion dependences *v(f)* in LiF crystals.

MATERIALS AND EXPERIMENTAL TECHNIQUES

This study addresses an important issue the influence of long-wave X-ray radiation of low doses on the frequency dependence of the elastic wave velocity, *v(f)*, in the frequency interval of 37.5…172.5 MHz in LiF crystals with the residual strain $\varepsilon = 0.4\%$ and at $T = 300$ K. The selector method, as sufficiently effective, was chosen by the authors to measure the velocity of the propagation of ultrasonic waves, and also the installation described in [5, 6] was used. The specimens with a purity of 10^{-4} wt.%, with the crystallographic orientation <100>, and 17x17x29 mm in dimension we used carrying out the experiments. According to the technology described in works [4–6, 8, 9], the specimens to study, after their cutoff, were finely polished to achieve the non-parallelism of working surfaces of approximately 1 μm/cm, which was controlled by means of an IKV optimeter. The surface non-parallelism 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 0.8 T_{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 $\varepsilon = 0.4\%$. 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 [4–6, 8, 9], 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, 6].

RESULTS AND DISCUSSION

Fig. 1 shows that the experimental dependences *v(f)* shown for the LiF crystals are not intense (curve 1) [5, 6] and are deformed and irradiated later with X-rays of 400 and 800 R (curves 2, 3). It can be seen that the nature of the frequency dependences *v(f)* varies significantly when the dislocation structure of the crystal passes from one state to another. Initially, when "growing" dislocations are fixed by the impurity atoms in a non-deformed (annealed) crystal, the propagation velocity of acoustic waves practically linearly changes when the frequency increased (curve 1).

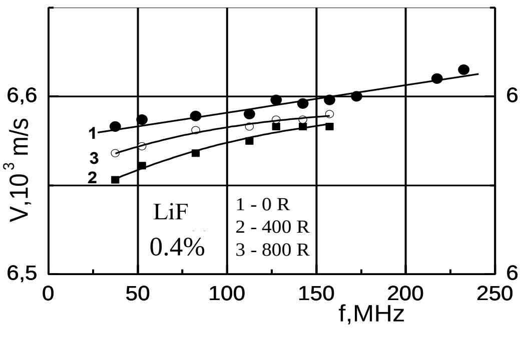

Fig. 1. Frequency dependences of the ultrasonic propagation velocity in LiF crystals where (1) a non-strained and non-irradiated specimen, (2, 3) a specimens with a residual deformation of 0.4%, and X-ray – irradiated to 400 and 800 R, respectively

Then, with the appearance of a significant number of highly mobile dislocations in the crystal owing to its deformation to 0.4% the frequency dependence *v(f)* emerged a pronounced interval with dispersion, which is especially notable in the low-frequency section. The X-irradiation of specimen to doses 400 and 800 R approached the frequency dependences *v(f)* (curves 2, 3) to the frequency position of curve 1 for the non-deformed specimen. The results presented in Fig. 1 can be interpreted in accordance with [7] in the following way. At low frequencies, the dislocation moves in an overlapping phase, and the actual hardness of the crystals is lower than that of non-dislocation. When the frequency increases, the synchronism between the dislocation moves and the excited external field is substantially violated, and the modulus of elasticity 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* (Fig. 2). We note that the modulus defect was calculated in this work using the equation $2\cdot\Delta V/V_\infty = \Delta C_{11}/C_{11}$, where $\Delta V = V_\infty - V$. The purely elastic velocity $V_\infty = 6.61\cdot10^3$ m/s was measured at a frequency of 217.5 MHz (see Fig.1, curve 1), when the crystal behaves itself as an ideal one owing to the absence of dislocation effects.

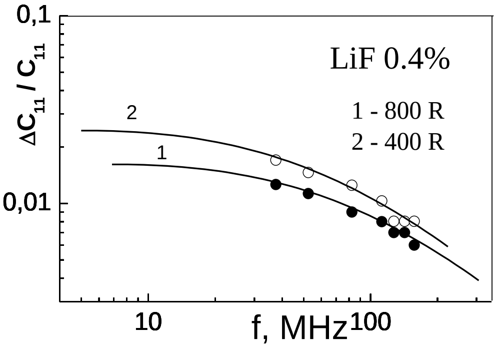

Fig. 2. Frequency dependences of the modulus defect in LiF crystals deformed to 0.4%, and X-ray-irradiated to 400 (2) and 800 R (1) respectively. Solid curves are the theoretical curves taken from work [10]

The Fig. 2 demonstrates that the experimental curves (1, 2) decrease by amplitude and are shifted toward higher frequencies with the increase of X-irradiation dose. The array of experimental points in Fig. 2 was approximated by the frequency dependence of the modulus defect calculated in work [10] 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 [5, 6]. 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.

It should be noted that the absolute values of ultrasound velocity in the frequency interval 37.5…172.5 MHz were measured to an accuracy of 0.05…0.1% [6], whereas the modulus defect $(\Delta C_{11}/C_{11})_0$ and the frequency were determined from the data (see Fig. 2) with an accuracy of 5…7 or 15…20%, respectively. On the basis of experimental results depicted in Fig. 2 and using the theoretical relations describing the low-frequency branch of the dislocation resonance [7], 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 [7], the formula for the modulus defect extrapolated to the low-frequency region looks like:

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

Substituting the expressions $\Delta_0 = \frac{8\cdot G\cdot b^2}{\pi^3\cdot C}$, $L^2 = \frac{0.084\cdot\pi\cdot C}{2\cdot B\cdot f_m^0}$, taken from work [7] into Eq. (1), we obtain a relation for the calculation of Λ in the form:

$$\Lambda = \frac{\pi^3\cdot f_m^0\cdot B_e}{2.016\cdot\Omega\cdot G\cdot b^2}\cdot\left(\frac{\Delta C_{11}}{C_{11}}\right)_0, \qquad (2)$$

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 $G\cdot b^2 = 28.77\cdot10^{-10}$ Pa/m^2 [9], and the damping constant $B_e = 3.62\cdot10^{-5}$ Pa·s obtained recently for the researched crystals in work [9], we obtain the $\Lambda = 1.44\cdot10^{11}$ m^{-2} (400 R) and $\Lambda = 1.5\cdot10^{11}$ m^{-2} (800 R). 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 [11]. At the same time, the average value $1.47\cdot10^{11}$ m^{-2} calculated by formula (2) raises some doubts, because it is by an order of magnitude larger that the corresponding value $\Lambda_e = 1.45\cdot10^{11}$ m^{-2} found directly by counting the etch pits on the crystal surface [9]. 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 [7] became possible by the formula:

$$L = \sqrt{\frac{0.084\cdot G\cdot b^2}{B_e\cdot f_m^0\cdot(1-\upsilon)}}, \qquad (3)$$

where ν is Poisson's ratio. Substituting $\nu = 0.27$ [5, 6] into formula (3), we calculated L. As was expected, an increase of the exposure dose gives rise to a monotonic decrease of the effective length of a dislocation segment, L, from $L = 9.98\cdot10^{-7}$ m (400 R) to $7.81\cdot10^{-7}$ m (800 R) owing to its fixation by radiation-induced defects. However, the initial value of L obtained before the irradiation was approximately 2, 3 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:

$$B = \frac{2.016\cdot\Omega\cdot G\cdot b^2\cdot\Lambda_e}{f_m^0\cdot\left(\frac{\Delta C_{11}}{C_{11}}\right)_0\cdot\pi^3}. \qquad (4)$$

The results of calculations by this formula show that the damping coefficient B for LiF crystals, similarly to what was obtained in works [8, 9], does not depend on the irradiation dose (~ $3.6\cdot10^{-6}$ Pa·s). 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, 9] by analyzing the descending branch of the dislocation resonance. The determined results agrees with the theoretical conclusions [2] that the dynamic dislocation damping 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 dose and a decrease of the dislocation mobility, which reveals 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 [9, 12, 13]. 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. 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 [7], 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.

CONCLUSIONS

The effect of long-wave X-ray irradiation in small doses on the dispersion of ultrasound velocity, $v(f)$, in LiF crystals with a residual deformation of 0.4%, was studied in the frequency range from 37.5 to 172.5 MHz and at $T = 300$ K. Preliminary deformation of the samples was found to result in a pronounced dispersion region on the $v(f)$ curve, which was linear in undeformed samples. It was found that the phenomenon of scattering is especially noticeable at low frequencies, when the scattering of the elastic energy of ultrasonic waves is due to the high mobility of long dislocation loops. It is established that the dislocation damping is constant B and the dislocation density Λ remains unchanged with increasing radiation dose. An analysis of the results shows that the absolute value of B is less, and the value of Λ is greater by an order of magnitude than the corresponding values of B_e and Λ_e obtained in the framework of the usual methods of high-frequency internal friction and selective etching of the surface of the crystal, respectively.

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ВЛИЯНИЕ РЕНТГЕНОВСКОГО ОБЛУЧЕНИЯ НА УПРУГИЕ, ДИНАМИЧЕСКИЕ И СТРУКТУРНЫЕ ХАРАКТЕРИСТИКИ ПРОДЕФОРМОВАННЫХ КРИСТАЛЛОВ LiF

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Импульсным методом в области частот 37,5…172,5 МГц при T = 300 К исследованы влияния предварительной деформации ε = 0,4% и рентгеновского облучения дозами 400 и 800 Р на ход частотных зависимостей скорости ультразвука $v(f)$ в кристаллах LiF. Установлено, что коэффициент динамической вязкости B и плотность дислокаций Λ с увеличением дозы облучения остаются неизменными. Выявлено, что абсолютное значение величины B в 10 раз меньше, а величины Λ – во столько же раз больше значений, которые дают наиболее корректные методы: высокочастотного внутреннего трения и прямого подсчета ямок травления соответственно.

ВПЛИВ РЕНТГЕНІВСЬКОГО ОПРОМІНЕННЯ НА ПРУЖНІ, ДИНАМІЧНІ І СТРУКТУРНІ ХАРАКТЕРИСТИКИ ПРОДЕФОРМОВАНИХ КРИСТАЛІВ LiF

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Імпульсним методом в області частот 37,5…172,5 МГц при T = 300 К досліджено вплив попередньої деформації ε = 0,4% і рентгенівського опромінення дозами 400 та 800 Р на хід частотних залежностей швидкості ультразвуку $v(f)$ у кристалах LiF. Встановлено, що коефіцієнт динамічної в'язкості B і густина дислокацій Λ зі зростанням дози опромінення залишаються незмінними. 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.