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REGULAR ARTICLE

Peculiarities of the Frequency Dependences of the Dislocation Decrement of Ultrasound Attenuation for Ionic Crystals with Radiation Defects and Impurities

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An analysis of frequency dependences of dislocation decrement of ultrasound attenuation $\Delta_d(f)$ behavior in KBr, LiF and NaCl crystals was performed in experiments with variable temperature T and dislocation density Λ of crystals, as well as with variations in dose of X-irradiation in the studied samples. It is shown that, unlike the experiments with variable T and Λ, where the high-frequency asymptotes of the $\Delta_d(f)$ curves were represented by families of straight lines, in the experiments with X-irradiated samples, a fundamental difference is observed – they merge into a single straight line. Within the framework of the key relationship of the Granato-Lücke theory, which describes the behavior of dynamic and structural characteristics based on the data of the post-resonance region of the curves $\Delta_d(f)$, obtained within the framework of the amplitude-independent internal friction method, the justification of the obtained results of experiments with the variables T, Λ and the X-irradiation dose was made. A prediction was made that the approximation of the Granato-Lücke formula, which describes the behavior of high-frequency asymptotes for irradiated samples, should also work for experiments with a variable impurity composition of the studied crystals. A comparison of the obtained $\Delta_d(f)$dependences on pure KCl and KCl + Ba crystals (10^{-2} wt. %) was made, and the predicted effect of high-frequency asymptotes merging into a solid line in experiments on crystals with variable impurity composition is proved. It is proved that the approximation of the formula of the Granato-Lücke theory describes the anomalous behavior of the high-frequency asymptotes of the $\Delta_d(f)$ curves for experiments with a variable concentration of any weak stoppers of the Friedel type.

Keywords: Dislocation decrement, Deformation, Irradiation, Dislocation, Coefficient of dynamic damping, Impurities.

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1. INTRODUCTION

Today, ionic crystals are widely used [1-9] in the elemental base of devices for various purposes, for example, optics (UV and IR optics), X-ray spectral analysis (as natural crystal monochromators), dosimetry (thermoluminescent dosimetry, detectors, scintillators), acoustics (delay lines for electrical signals), laser technology (lasers with a tuned wavelength on color centers) for control and diagnosis of processes in reactors, therefore the practical benefit of their physical characteristics studying is high. In addition, these crystals are considered as model ones and the main data set for constructing and testing fundamental theories in solid-state physics was obtained on them.

For studies of dynamic and structural characteristics of ionic crystals, the method of amplitude-independent internal friction is used within the framework of the Granato-Lücke dislocation theory [10] and the Alshitz-Indenbom theory of dynamic inhibition of dislocations [11]. On the basis of measurements of the frequency dependences of the dislocation absorption of ultrasound $\Delta_d(f)$ in the megahertz frequency range, results were obtained for a significant number of LHCs: KBr, KCl, CsI, LiF, and NaCl, they were considered in the review paper [12]. For practically all of the mentioned crystals, the conducted experiments were aimed at studying the influence of temperature in the range of 77-300 K and the influence of the density of dislocations Λ (or previous deformation ε) on the localization of curves $\Delta_d(f)$, establishing the corresponding dependences of the damping coefficient $B(T)$ and $B(\Lambda)$ and the average effective length of the dislocation segment $L(T)$ and $L(\Lambda)$, as well as determining the active phonon mechanisms of dislocations braking in the framework of the theory [11]. The only exceptions were LiF crystals, for which $B(\Lambda)$ and $L(\Lambda)$ studies were performed within the framework of theory [10] by different methods – to compare their effectiveness and informativeness, and for which, in particular, X-irradiated samples were used. The peculiarity of the frequency spectra $\Delta_d(f)$ obtained by us on X-irradiated LiF samples with different doses of irradiation was that the high-frequency asymptotes used to calculate the dynamic and structural characteristics of crystals within the framework of the theory [10], in contrast to experiments with a variable dislocation structure of the samples or a variable temperature of the experiment, merged into one line. Such a result was obtained for the second time. Earlier, Stern and Granato conducted similar studies on high-purity copper samples and recorded

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such an unusual effect for the first time, but they did not provide a proper explanation for it [12].

In [13], we made a preliminary attempt to explain the anomalous behavior of high-frequency asymptotes for X-irradiated samples within the framework of the theory [10]. Certain predictions were also made regarding possible similar results for experiments with crystals enriched with impurities. In work [14], the effect of asymptotes merging into one line was noticed again and was recorded for NaCl crystals with different doses of X-ray irradiation in the range of 0-600 R and a residual strain of 0.5 %. In addition, the work [15] was aimed directly at acoustic studies of dynamic and structural characteristics in KCl crystals with impurities and it can be the basis for identifying a new effect – the merging of high-frequency asymptotes in experiments with variable impurity concentrations.

In view of this, the purpose of this work is an overall comprehensive analysis of all the obtained results and an attempt to finally understand what causes the effect of merging into one line the high-frequency asymptotes of the frequency spectra of the dislocation absorption of ultrasound $\Delta_d(f)$ in acoustic experiments with irradiated crystals and crystals with a variable impurity composition.

2. EXPERIMENTS, RESULTS AND DISCUSSION

For all KBr, KCL, LiF and NaCl crystals, the results of which will be analyzed in this work, the technology of mechanical processing of samples, their annealing, control of geometric dimensions and plane-parallelism of working faces, choice of the mode of plastic deformation and metallization of sample surfaces, as well as the methodology for conducting of microstructural, low-temperature and acoustic studies and processing of results within the framework of theories [10] are described in detail in the relevant previous works mentioned in the review [12] and are not given separately here.

The only thing that should be noted here is that all the obtained results were obtained in the same scientific laboratory [12] using the same equipment, means and experimental technologies on crystals of the same series, so an error in the data analysis caused by the presence of these factors should not be expected.

Fig. 1 shows the effect of temperature in the range of 77-300 K on the localization of frequency spectra of the dislocation decrement of ultrasound absorption $\Delta_d(f)$ in KBr crystals with a residual strain value of $\varepsilon = 0.75$ % [12].

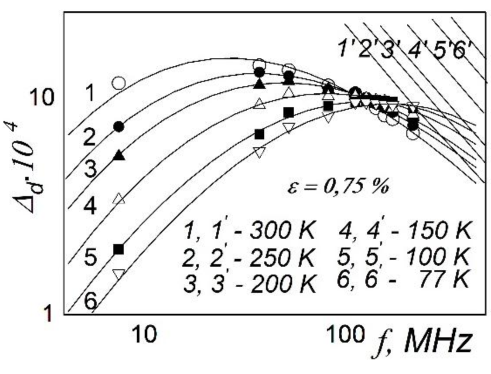

Fig. 1 – Frequency spectra $\Delta_d(f)$ in KBr crystals with $\varepsilon = 0.75$ % for temperatures 77-300 K

We can see that each of the curves $\Delta_d(f)$ has its own high-frequency asymptote. That is, we state that when the temperature is varied at a constant ε (or Λ), the asymptotes do not converge to one line.

Fig. 2 shows the influence of residual deformation for LiF samples in the range of 0.17-1.3 % at a temperature of $T = 300$ K on the localization of frequency dependences $\Delta_d(f)$ [12]. It can also be noted that the family of curves $\Delta_d(f)$ corresponds to a family of asymptotes. That is, we note that when varying ε (or the dislocation density Λ) at constant T, the coincidence of the asymptotes into one straight line is not observed.

It should be noted that the results presented for KBr and LiF demonstrate clearly defined patterns that apply to all studied LHCs [12] in studies with the variables T and Λ.

Now let's consider the results of studies of $\Delta_d(f)$, in which the experimental temperature and mechanical state of the material were fixed, but the dose of X-ray irradiation of the samples was varied.

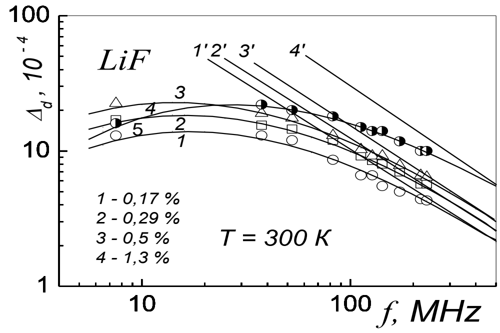

Fig. 2 – Frequency spectra $\Delta_d(f)$ in KBr crystals with values of residual deformation ε: 1 – 0.17 %, 2 – 0.29 %, 3 – 0.5 %, 4 – 1.3 % at $T = 300$ K

Fig. 3 shows the effect of the irradiation time (the X-irradiation dose rate at the location of the crystals was 0.11 R/s) on the localization of the frequency dependences $\Delta_d(f)$ in LiF crystals with a residual deformation $\varepsilon = 1$ % at a temperature $T = 300$ K [12]. The nature of radiation defects that arose during X-ray irradiation in LiF crystals was studied by us within the framework of the optical absorption method [16-19] in a separate paper [20].

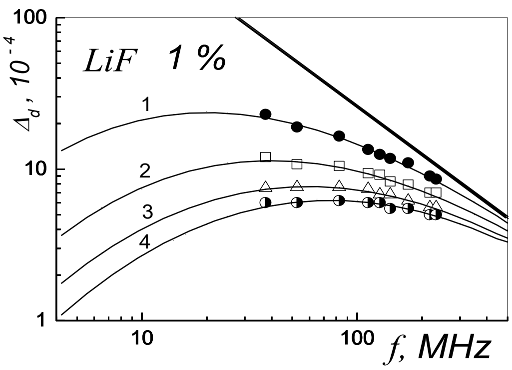

Fig. 3 – Frequency spectra $\Delta_d(f)$ for LiF crystals with $\varepsilon = 1$ % at $T = 300$ K for the irradiation time t, min.: 1 – 0; 2 – 20; 3 – 40; 4 – 60 minutes

Fig. 3 has a fundamental difference from the data presented in Fig. 1 and Fig. 2 – the high-frequency asymptotes of different frequency spectra represent a single common straight line.

A similar result was obtained for NaCl crystals [14], as shown in Fig. 4.

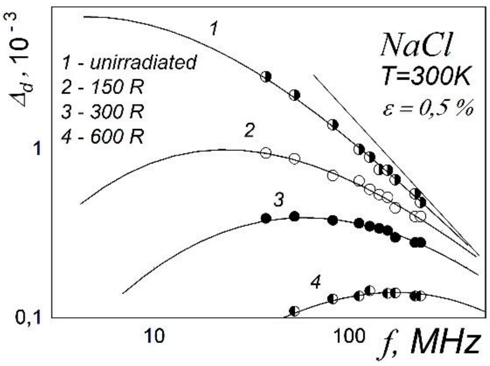

Fig. 4 – Frequency spectra $\Delta_d(f)$ for NaCl crystals with $\varepsilon = 0.5$ % at $T = 300$ K for samples with different doses of X-ray radiation

So, we can state that the results obtained earlier by Stern and Granato for Cu and by our group for LiF and NaCl clearly indicate that in experiments on samples with a constant dislocation density Λ (or ε) and a fixed temperature T, when the X-irradiation dose is varied, a stable pattern is observed – the frequency spectra of the dislocation decrement of ultrasound attenuation are described by a single high-frequency asymptote.

To substantiate this pattern, we propose to use the key relation of the Granato-Lücke theory, which describes the main method of determining the coefficient of dynamic braking of dislocations B from the post-resonance region of the measured dependences $\Delta_d(f)$ [10]:

$$B = \frac{4\Omega G b^2 \Lambda}{\pi^2 \Delta_\infty f_\infty}, \tag{1}$$

where Δ_∞ – decrement value for frequencies f_∞ significantly larger than the resonant one, Ω – orientation factor, G – shear modulus of the operating sliding system, b – magnitude of the Burgers vector.

Combination of parameters $\Omega \cdot G \cdot b^2$ is temperature dependent and independent of the dislocation density. In experiments with fixed T and variable Λ, as we know [12, 21], B does not depend on Λ, therefore the "equilibrium" of B is ensured by a combination of three parameters:

$$B = const \approx \frac{\Lambda}{\Delta_\infty f_\infty}, \tag{2}$$

The presence of a variable numerator also explains the asynchronous change of parameters in the combination $f_\infty \cdot \Delta_\infty$ when changing Λ, therefore Fig. 2 demonstrates a noticeable spread of high-frequency asymptotes, due to the fact that the equilibrium B is provided by three variable parameters.

In experiments with a variable T, the situation is even more complicated, because the damping coefficient depends on the temperature [11, 12] and this change is described by the behavior of the parameters of formula (1), which depend on T:

$$B \neq const \approx \frac{\Omega G b^2}{\Delta_\infty f_\infty}, \tag{3}$$

The presence of many parameters that control the process of temperature change in B causes the spread of high-frequency asymptotes, which is demonstrated by the results shown in Fig. 1.

Finally, let's consider an experiment with T = const, Λ = const (experiments with X-rayed samples). It is known that B does not depend on the radiation dose [12, 22], therefore, for this case, formula (1) acquires its "simplest" approximation:

$$B = const \approx \frac{1}{\Delta_\infty f_\infty}, \tag{4}$$

To ensure the invariance of B, the parameters f_∞ and Δ_∞ (which usually respond to a change in the structural state of the material, i.e., to the appearance of radiation defects) must change synchronously, and in such a way that their combination $f_\infty \cdot \Delta_\infty$ is constant. This means that the high-frequency asymptotes will represent one common line for all $\Delta_d(f)$ curves, which is demonstrated by the dependencies shown in Fig. 3, 4.

The approximation (4) presented by us not only substantiates the regularity of acoustic experiments with X-ray irradiated samples, but also allows us to make other assumptions.

It is obvious that not only radiation defects as point centers of Friedel type pinning should lead to the convergence of asymptotes into one solid line. Here we can mention other "weak" stoppers on Friedel-type dislocations [10], for example, impurities or magnetically sensitive stoppers.

If this is indeed the case, then approximation (4) should also explain the experiments with adding of additives into crystals at T = const, Λ = const. To test this assumption, we considered the results of our recent work on KCl [15].

Fig. 5 shows the effect of residual deformation of KCl + Ba samples (10^{-2} wt. %) in the range of 0.25-0.75 % at a temperature of T = 300 K on the localization of frequency dependences $\Delta_d(f)$ [15].

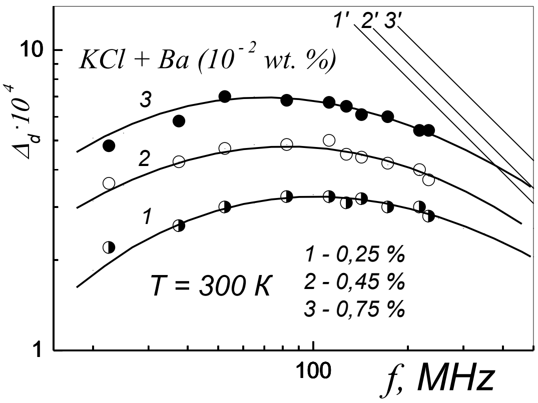

Fig. 5. – Frequency spectra $\Delta_d(f)$ in crystals KCl + Ba (10^{-2} wt. %) with different values of ε at $T = 300$ K

It is seen that the curves $\Delta_d(f)$ have different high-frequency asymptotes. It is explained by the approximation described by formula (2) for experiments with fixed T and variable Λ. From these curves, within the framework of the Granato-Lücke theory, calculations of the dynamic braking coefficient of dislocations B were performed, the results of which are shown in Fig. 6.

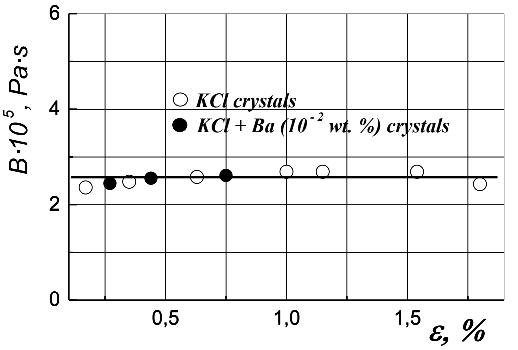

Fig. 6 – Dependence B (ε) for KCl + Ba crystals (10^{-2} wt. %) and conditionally pure KCl crystals

The results presented in Fig. 6 demonstrated for the first time that the value of B does not depend on the purity of the crystals (as well as on the presence of radiation defects and on the dislocations density), which confirms the theoretical predictions of Alschitz-Indenbohm [11] that the level of B is limited only by the interaction of mobile dislocations with the phonon subsystem of crystals.

Within the framework of our work, the results presented in Fig. 6 are interesting for us in the sense that if the value B is constant, then at T = const, Λ = const, approximation (4) should work and the high-frequency asymptotes of the dependences $\Delta_d(f)$ for conditionally pure KCl crystals and KCl + Ba (10^{-2} wt. %) crystals deformed to the same value ε should coincide. To test this assumption, we used two frequency spectra with the same $\varepsilon = 0.45$ % – from previously obtained data for pure KCl crystals [12] and for KCl + Ba (10^{-2} wt. %) samples [15], which are presented in Fig. 7.

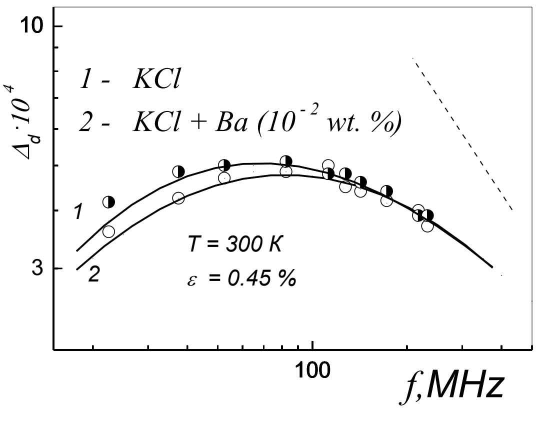

Fig. 7. – Dependences $\Delta_d(f)$ for KCl + Ba crystals (10^{-2} wt. %) and conditionally pure KCl crystals with $\varepsilon = 0.45$ % at $T = 300$ K

It can be seen from Fig. 7, that the frequency spectra $\Delta_d(f)$ have a common high-frequency asymptote because the conditions of relation (4) are satisfied (B = const (see Fig. 6), T = const, Λ = const).

Thus, as a result of the work, we have found out the reasons for the atypical behavior of the frequency spectra of dislocation absorption of ultrasound in X-ray irradiated crystals in terms of their description by a fixed high-frequency asymptote at a fixed temperature and dislocation density, and, in addition, we have made a prediction and verified that it is fulfilled for acoustic experiments with crystals in which pinning centers are introduced on Friedel type dislocations of any physical nature under the condition T = const, Λ = const.

3. CONCLUSIONS

1. In this work, based on a systematic consideration of the behavior of the frequency spectra of the dislocation decrement of ultrasound attenuation in a number of ionic crystals – KCl, KBr, LiF and NaCl – the influence of temperature, dislocation density, X-ray dose and the presence of impurities on the localization of the $\Delta_d(f)$ dependences and the behavior of their high-frequency asymptotes, which are used to calculate the dynamic and structural characteristics of crystals within the framework of the Granato-Lücke theory, is analyzed.

2. Based on the analysis of the parameters included in the basic calculation relation of the Granato-Lücke theory for amplitude-independent internal friction, approximations are considered that sufficiently accurately describe the atypical behavior of high-frequency asymptotes in acoustic experiments with X-ray irradiated crystals.

3. For the first time, a theoretical justification is provided for the experimental regularities for $\Delta_d(f)$ in experiments where the experimental temperature and the dislocation density of the samples remain unchanged, and only the number of weak pinning centers – radiation defects on the dislocations – changes.

4. It is predicted that the effect of merging of high-frequency asymptotes into one line can be observed not only for X-ray irradiated samples, but also for crystals into which Friedel pinning centers on dislocations of any other physical nature (impurities, magnetically sensitive stoppers) are implanted.

5. Based on a comparison of the data obtained by us on $\Delta_d(f)$ for KCl + Ba crystals (10^{-2} wt. %) and conditionally pure KCl crystals with the same value of $\varepsilon = 0.45$ % at a fixed experimental temperature $T = 300$ K, it was shown that the high-frequency asymptotes for the $\Delta_d(f)$ spectra represent a common line, which confirms the validity of the analytical justification carried out in this work and the theoretical predictions made on its basis.

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Особливості частотних залежностей дислокаційного декременту затухання ультразвуку для іонних кристалів з радіаційними дефектами і домішками

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Проведено аналіз поведінки частотних залежностей дислокаційного декременту затухання ультразвуку $\Delta_d(f)$ в кристалах KBr, LiF і NaCl в дослідах зі змінною температурою T та густиною дислокацій Λ кристалів, а також з варіюванням дози х-опромінення у досліджуваних зразках. Показано, що на відміну від дослідів зі змінними T і Λ, де високочастотні асимптоти кривих $\Delta_d(f)$ були представлені сімействами прямих ліній, в дослідах з Х-опроміненими зразками спостерігається докорінна відмінність – вони зливаються в єдину пряму. В рамках ключового співвідношення теорії Гранато-Люкке, що описує поведінку динамічних і структурних характеристик на основі даних після резонансної області кривих $\Delta_d(f)$, одержаних в рамках методу амплітудно-незалежного внутрішнього тертя, зроблене обґрунтування отриманих результатів експериментів зі змінними T, Λ і дозою Х-опромінення. Зроблене передбачення, що наближення формули Гранато-Люкке, що описує поведінку високочастотних асимптот для опромінених зразків має спрацьовувати і для експериментів зі змінним домішковим складом досліджуваних кристалів. Проаналізовано роботи на чистих кристалах KCl і KCl + Ba (10^{-2} ваг. %), зроблене зіставлення отриманих залежностей $\Delta_d(f)$ і, таким чином, доведено передбачуваний нами ефект злиття високочастотних асимптот в суцільну лінію в експериментах на кристалах зі змінним домішковим складом. Доведено, що наближення формули теорії Гранато-Люкке успішно описує аномальну поведінку високочастотних асимптот кривих $\Delta_d(f)$ як для дослідів зі змінною дозою Х-опромінення в зразках, так і в експериментах, де варіюється концентрація домішок в кристалах. Зроблено висновок, що аномальна поведінка високочастотних асимптот кривих $\Delta_d(f)$ є абсолютно передбачуваною для дослідів зі змінною концентрацією будь яких слабких стопорів фриделівського типу.

Ключові слова: Дислокаційний декремент, Деформація, Опромінення, Дислокації, Коефіцієнт динамічного гальмування, Домішки.