DETERMINATION OF THE DISPERSION OF ELASTIC VIBRATIONS IN THE CRYSTAL LATTICE OF POTASSIUM CHLORIDE FROM DIFFUSE X-RAY SCATTERING

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DETERMINATION OF THE DISPERSION OF ELASTIC VIBRATIONS IN THE CRYSTAL LATTICE OF POTASSIUM CHLORIDE FROM DIFFUSE X-RAY SCATTERING

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Determination of the dynamic characteristics of crystals (for example, the spectrum of thermal vibrations and elastic constants) from diffuse X-ray scattering has a number of advantages in comparison with other known methods (measurement of heat capacity, mechanical and ultrasonic methods), since this method makes it possible to determine not values of the measured quantities averaged over the whole crystal, but their local values. In some cases this proves especially important. Thus, for example, under plastic deformation of crystals, distortions of the crystal lattice are local in character, and it should be expected that the elastic constants and other quantities associated with them will differ greatly from one place in the crystal, where there is a high density of dislocations, to another place, where there are few of them. This is indicated by the sharp difference in Debye temperatures, determined by the X-ray method, in the region of the slip band and outside it ([1]).

The method of diffuse X-ray scattering has so far been used only for the experimental determination of the spectrum of thermal vibrations in metals and in some metallic solid solutions. By this method frequency spectra have been found for Cu ([2]), Al ([3]), α-Fe ([4]), Ni ([5]), and a number of other substances. In the case of alkali-halide crystals the lattice is formed by ions of two kinds with different masses, which leads to the appearance of two branches of elastic vibrations—the optical and the acoustic. In determining the spectrum of thermal vibrations from diffuse X-ray scattering, it is difficult to separate the scattering due to the acoustic and optical branches. Evidently this explains why, for the determination of elastic vibrations in alkali-halide crystals, with the exception of AgCl ([6]), the diffuse-scattering method has not yet been applied.

However, there are several alkali-halide crystals (NaF, KCl, RbBr, and CsJ) in which the atomic numbers and, consequently, the atomic scattering factors and the masses of the ions differ very little from one another. Therefore, in a first approximation the polarization vectors of the acoustic and optical branches of the crystal lattices of such substances may be regarded as equal. Then, owing to the monoatomic nature of the model, the acoustic branch passes continuously into the optical one, forming "quasiacoustic" branches. Therefore, in these cases one can use the techniques applied to determine elastic vibrations in the crystal lattices of metals from diffuse X-ray scattering.

In the present work the results of an investigation of diffuse X-ray scattering in undeformed potassium chloride crystals are described.

Measurement of the angular distribution of the intensity of diffuse X-ray scattering in principle makes it possible to determine the spectrum of elastic vibrations. However, it turns out that, in order to obtain the curve of the frequency spectrum, it is necessary to solve a secular determinant for 3000–4000 wave vectors. Therefore, in some works (^{5}) one is limited to obtaining—

···by the construction of dispersion curves, i.e., the dependence of the vibration frequency on the wave vector, which is also of interest, if only because it makes it possible to determine the elastic constants of the crystal. In the present work, too, only the dispersion curves were determined, and from them the elastic constants.

To construct the curves, the frequency—the magnitude of the wave vector—was found using the well-known relation of James (^{7}), which connects the intensity of first-order diffuse X-ray scattering with the frequency

$$I_T = NIef^2e^{-2M}\left|\frac{\mathbf{s}}{\mathbf{k}}\right|^2 \sum_{j=1}^{3} \frac{\cos^2(\mathbf{s}, \mathbf{e}_{kj})}{\nu_{kj}} \left[\frac{1}{e^{h\nu_{kj}/kT} - 1} + \frac{1}{2}\right], \qquad (1)$$

where $|\mathbf{s}|$ is a reciprocal-lattice vector, $|\mathbf{k}|$ is the wave vector, and ν_{kj} is the frequency for the given $\mathbf{k}$ of the j-th polarization (the remaining notation is generally accepted).

However, this relation is greatly simplified if measurements are carried out for directions in which the propagating waves have a purely longitudinal or transverse component. For cubic crystals, to which the potassium chloride crystal belongs, such directions are [100], [110], [111].

The experimentally measured intensity contains scattering of the II and III orders, as well as Compton scattering. The Compton scattering was determined by calculation. Scattering of the II and III orders was not taken into account, which introduced an error in the determination of the intensity of up to 20%.

The specimens for the investigation were obtained by cleavage along cleavage planes or by cutting with a wet thread from a massive potassium chloride single crystal grown by the Kyropoulos method. The specimens were then annealed for 8-10 hours at 700°. The dislocation density in such specimens did not exceed 10^4 cm^{-2}.

Measurements of the diffuse-scattering intensity were carried out on a URS-50I apparatus using radiation from a copper anode at a voltage of 30 kV and a tube current of 7 mA. The radiation was monochromatized by a bent lithium fluoride crystal. The divergence of the beam was limited vertically and horizontally by means of a set of slits and collimators. From the total intensity recorded by the counter, the intensity of scattering by air (background) was subtracted. The result obtained was then converted into absolute units by comparison with the scattering by paraffin at large angles.

Intensity measurements were carried out between the nodes 200–400, 200–220, 222–444, 222–402, 220–440, 220–400, 220–222. Each point of reciprocal space received 3000 pulses. In the calculations an "oblique" correction (^{8}) and a correction for Compton scattering were introduced. After allowing for the corrections, the frequencies of longitudinal and transverse waves were calculated from formula (1), and the dependence of these frequencies on the wave vectors was constructed. The dispersion curves for longitudinal and transverse waves in the directions [100], [110], and [111] are given in Fig. 1.

Table 1

$C_{11} \cdot 10^{-11}$, dyn/cm^2	$C_{12} \cdot 10^{-11}$, dyn/cm^2	$C_{44} \cdot 10^{-11}$, dyn/cm^2	Source
3.5	0.98	0.68	Data obtained in the present investigation from the dispersion curves
3.85	0.75	0.79	Wooster (^{8}); data obtained by the photographic method from diffuse X-ray scattering
4.095	0.705	0.63	Lazarus (^{9}); data obtained by the ultrasonic method

The slope of the dispersion curves at small values of the wave vector characterizes the relation between the elastic constants (C_{11}, C_{12}, C_{44}). The corresponding values of the elastic constants were calculated (Table 1).

Fig. 1

Figure 1: Fig. 1

As follows from the results of measurements by the ultrasonic method, the values of the elastic constants of alkali-halide crystals for different specimens obtained from a single crystal may differ from one another by 8-10%. In the present work the elastic constants obtained from diffuse scattering of X-rays, despite the neglect of second-order scattering, are in good agreement with the values obtained from ultrasonic measurements.

Fig. 1. Dispersion curves of elastic waves propagating along the axes [100] KCl (*I*), [110] KCl (*II*) and [111] KCl (*III*). L—longitudinal waves; T—transverse waves; T_1—transverse waves, polarization vector parallel to [001]; T_2—transverse waves, polarization vector parallel to $[1\bar{1}0]$.
a—results of the present work; *b*—results of the Tolpygo model; *v*—results of the Kucher model.

Table 1 also gives the values of the elastic constants determined by Wooster by a photographic method from a single X-ray photograph. These results likewise agree well with the results of the present work.

The dispersion curves for KCl have been calculated theoretically by a number of authors ($^{10-12}$), who proceeded from different models. Iona (^{10}) calculated the frequencies of KCl for a number of wave vectors, proceeding from a model of point atoms of identical mass. Tolpygo and Zaslavskaya (^{11}) and Kucher (^{12}) took into account the deformation of the electronic shells of the ions due to interaction with the electric fields in the lattice and also determined the frequencies as a function of the wave vector for potassium chloride. A comparison of the experimental values obtained in the present work with the calculated data in the works of Tolpygo and Kucher (Fig. 1) shows that for the directions [100] and [110] they are in good agreement. The experimental curves for the [111] direction lie below the calculated values. This is probably due to the fact that it is very difficult to prepare an artificial (111) surface without introducing distortions into it. These distortions lead to a change in the intensity of X-ray scattering, as a result of which the dispersion curves obtained no longer correspond to a perfect crystal lattice. However, this circumstance does not affect the slope of the curves near the origin of coordinates, from which the elastic constants are determined.

Iona’ s data are not shown in the figure. They differ strongly both from the experimental results and from the calculated curves of Tolpygo and Kucher. Evidently, Iona’ s approximation is very crude, and in calculating dispersion curves one cannot neglect the deformability of the electronic shells of the ions.

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