SCATTERING OF LIGHT IN THE PHASE TRANSITION OF THE NH_4Cl CRYSTAL

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The intense Rayleigh scattering of light in a crystal undergoing a phase transition was observed and investigated so far only in quartz [1,2]. The phenomenon is observed at 573°C, when the quartz undergoes a second-order phase transition close to the Curie point.

An interesting object for further research on light scattering during phase transtitions of crystals is NH_4Cl. The unique phase transition of this crystal at -30°C has been thoroughly investigated by various methods [3,4]. It has been established that this is apparently a first-order transition, but with a low transition heat. The latter circumstance is of particular interest, since it may explain whether the intense light scattering observed in quartz is an exclusive feature of second-order phase transitions. We have assumed that an intensification of light scattering, albeit to a lesser degree, should take place also in first-order phase transitions if the crystal transition heat is small and if the statistical fluctuations of the crystal-lattice parameters can be appreciable near the phase transition region. Of course, for light scattering it is also necessary that these fluctuations produce considerable inhomogeneities of the refractive index of the crystal, something not necessarily connected with any rearrangement of the crystal lattice.

The general scheme of our experiment with NH_4Cl single crystals is similar to that described in [1,2]. A block of NH_4Cl cut parallel to the edges of the unit-cell cube of the crystal lattice was placed in a cryostat. The primary light beam was parallel to one of the edges of the cubic lattice. The electric vector in the primary light beam was perpendicular to the scattering plane. A photomultiplier was used to measure the intensity of the light, scattered by the crystal at 90° to the primary light beam, through the sighting window of the cryostat. We made two simultaneous series of observations by this method.

In the first series of measurements the scattered light incident on the photomultiplier had the same linear polarization as the primary light beam. It follows from the complete theory of Rayleigh scattering of light in crystals, developed by Motulevich [5], that this series of experiments could yield a record of the scattering of light by longitudinal Debye thermal waves propagating along the diagonal of the cube face. The results of the first series of experiments are shown in Fig. 1. The abscissas represent the temperature measured from the temperature corresponding to the maximum intensity of the scattered light; the ordinates represent the intensity of the scattered light in relative units. Different designations of the experimental points on the diagram correspond to different experiments and to different wavelengths. In the second series of experiments the photomultiplier received scattered light with an electric vector in the plane of incidence. In this case, as follows from Motulevich's paper, we could observe light scattered by transverse Debye thermal waves propagating along the diagonal of the crystal-cube face and polarized perpendicular to the

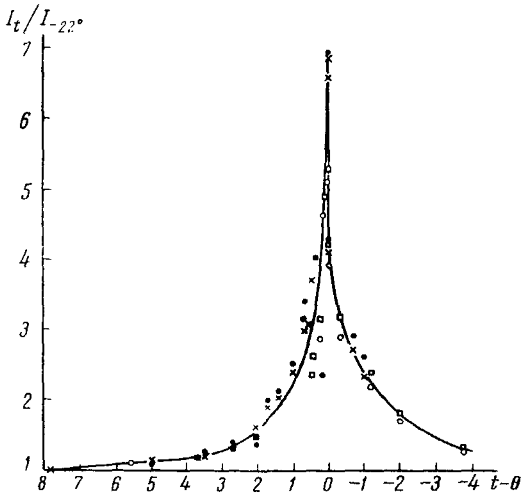

Fig. 1

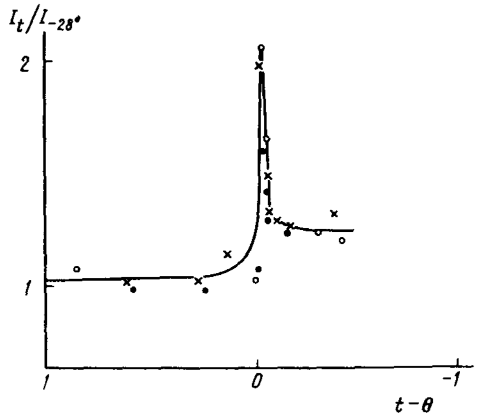

Fig. 2

light-scattering plane. The results of the second series of experiments are shown in Fig. 2. In both series of experiments the measurements were made on the 4358 and 5460 Å mercury lines.

The results of both series of experiments demonstrate convincingly the presence of the expected effect, viz., intensification of the scattering of light during the phase transition of the NH_4Cl crystal. The figures show clearly several differences in the temperature dependence of the intensity of scattered light of differing polarization. It must also be pointed out that the maxima of the scattered-light intensity in the first and second series of experiments occurred at somewhat different temperatures ($\Delta T \sim 0.02°$). The maximum for the component perpendicular to the scattering plane occurs at a lower temperature.

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