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X-Ray Scattering from the Displacement Field of Point Defects*

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We measured diffuse x-ray scattering from the displacement field of point defects in KBr single crystals. From these results absolute values for the double-force tensors of the anion vacancy and the interstitial anion can be derived. From the symmetry of the displacement field a model for the interstitial anion is proposed.

Diffuse x-ray scattering from crystals containing a random distribution of point defects which cause a displacement field was considered theoretically long ago.[1-7] Only recently has the diffuse x-ray scattering from crystals with defect clusters been observed experimentally.[8-12] This has initiated further theoretical work.[13-16]

In this Letter we report measurements of the diffuse x-ray scattering close to Bragg peaks due to point defects [Huang diffuse scattering (HDS)] in low-temperature x-irradiated KBr crystals.[17] In spite of the large amount of information about the nature and concentration of radiation-induced defects in KBr, the structure of the interstitial anion is unknown. HDS gives information about the displacement field of a defect. In favorable cases one can deduce the structure of the defect itself. We have measured the intensity distribution of HDS in certain directions of reciprocal space. From the absolute scattering intensity, elastic double-force tensors were determined describing the long-range displacement fields of the interstitial anion and the anion vacancy in KBr.

In the following we keep close to the formulation of the theory given recently by Trinkhaus,[16] which is best suited to evaluate experimental results. The displacement field of a point defect in a solid can be described by a double-force tensor P_{ij}. The intensity distribution $I((hkl),[hkl])$ in the direction $[hkl]$ close to a reciprocal lattice point (hkl) gives the quadratic double-force–tensor quantities α,β,γ, which for cubic crystals with defects, statistically distributed over possible orientations, are given by

$$\alpha=\sum_i P_{ii}^2,\quad \beta=\sum_{i>j} P_{ii}P_{jj},\quad \gamma=\sum_{i>j} P_{ij}^2. \tag{1}$$

For the reciprocal-lattice points and directions of our experiment one gets

$$I_H((600),[100])=I\alpha/C_{11}^2, \tag{2}$$

$$I_H((440),[110])=2I\,\frac{\alpha+\beta+2\gamma}{(C_{11}+C_{12}+2C_{44})^2}, \tag{3}$$

$$I_H((440),[1\bar{1}0'])=2I\,\frac{\alpha-\beta}{(C_{11}-C_{12})^2}, \tag{4}$$

$$I_H((440),[001])=I_H((600),[010])=I\gamma/C_{44}^2, \tag{5}$$

$$I=(n|F|^2/3v^2)(K/g)^2. \tag{6}$$

n is the concentration of defects, $\vec{K}=\vec{k}-\vec{k}_0$ is the scattering vector, $\vec{g}=\vec{K}-\vec{G}$, $\vec{G}$ is the reciprocal-lattice vector of the Bragg peak, v is the volume of the unit cell, $|F|$ is the scattering amplitude of the unit cell at the measuring temperature,[18] and C_{11}, C_{12}, C_{44} are elastic constants.[19]

The experimental setup will be described in detail elsewhere. The intensity distribution of scattered Cu $K\alpha_1$ radiation having passed a bent quartz monochromator was measured point by point for various settings of crystals and detector angle. The incoming intensity was monitored by a second proportional counter. Intensity was put on an absolute scale in the usual way by scattering from polystyrene.[20] The defect-induced

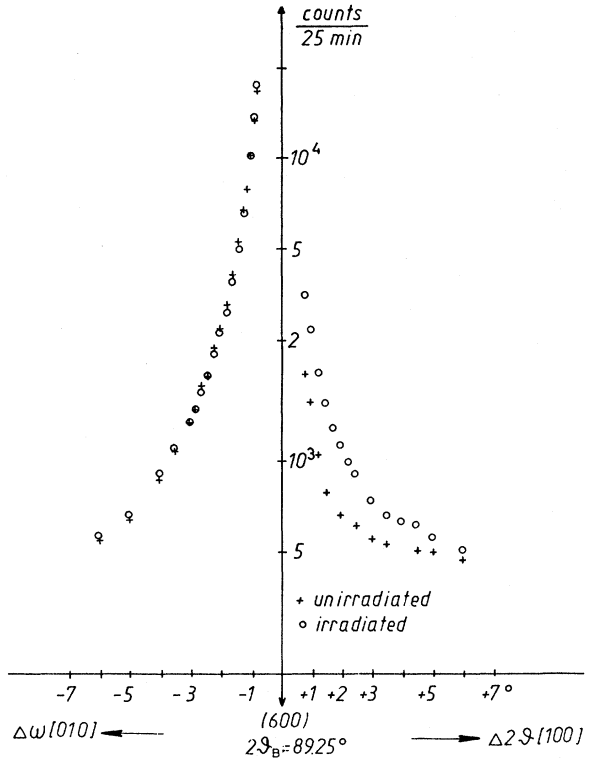

FIG. 1. Diffuse scattered x-ray intensity distribution of a KBr single crystal at 6 K close to the (600) reciprocal-lattice point in the [100] and [010] directions before and after x-irradiation.

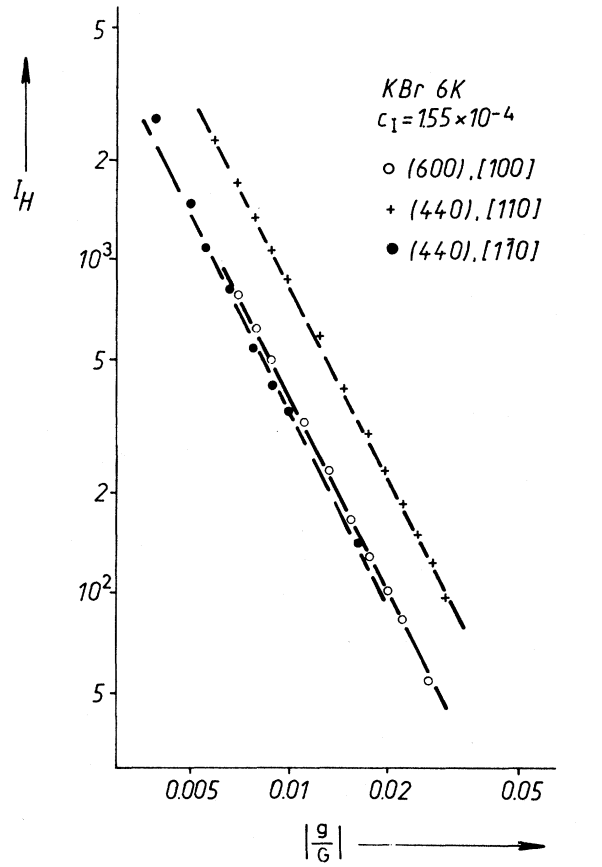

FIG. 2. Huang diffuse-scattering intensity (in electron units) as a function of relative distance from reciprocal-lattice points.

HDS intensity was obtained as the difference between scattered intensity from the crystals after and before x irradiation. Pure KBr single crystals (Harshaw) were used. Defects were created by x irradiation (100 kV, 18 mA, tungsten anode tube) at 6 K. The crystals were kept at 6 K in a cryostat[21] to avoid thermal annealing of the defects and to keep thermal diffuse scattering low. Defect concentration was determined from simultaneously measuring the length change of the crystals. Balzer's[22] results were used to determine the actual concentrations of defects from the length-change data.

Figure 1 shows a typical result; $I((600),[100])$ and $I((600),[010])$ were measured for a crystal before and after irradiation. Measurements were performed in positive and negative directions of $\vec{g}$. In Fig. 1 the average values from both measurements are given. Most remarkably, $I_H((600),[010])$ is zero. This gives $\gamma \approx 0$ immediately according to Eq. (5).

In Fig. 2 the HDS is plotted as a function of g on a double logarithmic scale. The predicted $1/g^2$ dependence [Eq. (6)], given by the dashed lines, is followed over the whole range.

From the measurement of $I_H((440),[001])$, $\gamma \approx 0$ was confirmed. The measurements of $I_H((600),[100])$, $I_H((440),[110])$, and $I_H((440),[1\bar{1}0])$ give, together with Eqs. (2) to (6),

$$\alpha = 410 \pm 27 \text{ eV}^2, \quad \alpha + \beta = 711 \pm 55 \text{ eV}^2,$$
$$\alpha - \beta = 115 \pm 8 \text{ eV}^2, \quad \gamma \leq 0.4 \text{ eV}^2.$$

During low-temperature irradiation two types of Frenkel pairs are created[22]: (1) $F+H$ centers; their double-force tensors are known.[19,23] Therefore, their contribution to α, β, and γ can be taken into account. (2) $\alpha+I$ centers, i.e., anion vacancies and interstitial anions. The volume change by this pair, which is connected with the trace of P_{ij}, has been determined.[22] Using this result and the fact that the anion vacancy has cubic symmetry we get the following results: For the *anion vacancy* the volume change $\Delta v = 0.7 \pm 0.2$ atomic volumes, and the double-force tensor $P_{ij} = \delta_{ij}(2.8 \pm 0.6)$ eV; for the *interstitial*

anion the volume change $\Delta v = 2.5 \pm 0.2$ atomic volumes, and two values for the double-force tensor are possible,

$$P_I = \begin{pmatrix} 6.9 \pm 1.9 & 0 & 0 \\ 0 & 6.9 \pm 1.9 & 0 \\ 0 & 0 & 16.3 \pm 3.8 \end{pmatrix} \text{eV}$$

or

$$P_I' = \begin{pmatrix} 13.1 \pm 3.1 & 0 & 0 \\ 0 & 13.1 \pm 3.1 & 0 \\ 0 & 0 & 3.8 \pm 1.9 \end{pmatrix} \text{eV}.$$

The intensity distribution in reciprocal space clearly shows that the defects observed here are cube-edge oriented. Cube-edge–oriented defects with all diagonal elements of the double-force tensor different from each other are very unlikely. Cubic symmetry can be excluded from $\alpha - \beta \neq 0$. Therefore, we have to assume that the displacement fields have tetragonal symmetry.

Only such locations of the interstitial ion within the elementary cell may be considered which cause a displacement field of tetragonal or at least cube-edge–oriented symmetry. The simplest model for the interstitial anion which is consistent with this would be a pair of *two* bromine ions on *one* anion lattice site, with internuclear axes in ⟨100⟩ directions. In this case P_I would be most probably the corresponding double-force tensor.

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