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Résumé. — La création des défauts anisotropiques comme les centres H et V_K dans les halogénures alcalins conduit à une contorsion anisotropique du cristal. La rotation de ces défauts d'une orientation à une autre par irradiation avec de la lumière polarisée conduit à un changement des dimensions macroscopiques du cristal. Avec les mesures d'absorption optique, le tenseur de force double, qui décrit l'interaction mécanique du défaut avec le réseau moléculaire, est obtenu.

Abstract. — The creation of anisotropic defects as H- and V_K-centers in alkalihalides leads to an anisotropic distortion of the crystal. Rotation of these defects from one orientation to another by irradiation with polarized light leads to a change of the macroscopic dimensions of the crystal. Together with optical absorption measurements the double force tensor describing the mechanical interaction of the defect with the lattice is obtained.

H- and V_K-centers in alkalihalides are optically and mechanically anisotropic. Their creation involves an anisotropic distortion of the crystal. Anisotropic defects usually have different equivalent orientations in the lattice.

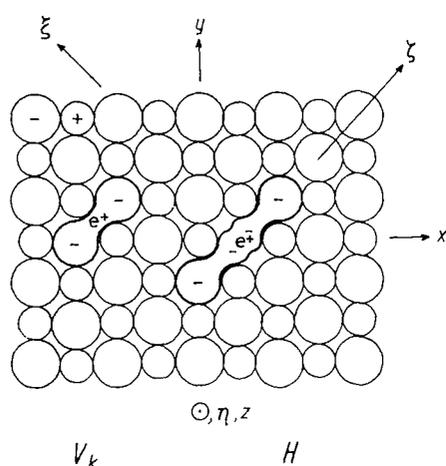Figure 1 shows the models of H- and V_K-centers. The H-center is an interstitial atom (Cl_2^--molecule

FIG. 1. — H- and V_K-centers in alkali halides.

on one lattice site), and the V_K-center is a trapped hole center (Cl_2^--molecule on two lattice sites). Both centers are well known from ESR and optical measurements. The optical and the elastic dipole axes are along $\langle 110 \rangle$ directions. The coordinates ξ, η, ζ

are the principal axes of the defect, and the coordinates x, y, z are the usual crystal coordinates.

Defects in alkalihalides lead to characteristic absorption bands in the otherwise transparent range between about 0.1 and 6 eV. Information about the species and concentration of the different defects can be obtained relatively easily by optical absorption measurements.

Due to the high symmetry of the cubic lattice there exist 6 equivalent orientations for a $\langle 110 \rangle$ oriented defect.

If uniaxial stress is applied to the crystal, the orientationally degenerated levels split up. The population of the now energetically different levels obeys a Boltzmann distribution. The concentration of the defects of different orientations as a function of applied stress can be measured as optical absorption. This gives information about the anisotropic part of the double force tensor.

A new method will be described here. It takes advantage of the possibility to rotate these anisotropic defects by polarized light. The anisotropic defects considered here have a well defined reorientation temperature. Above this temperature defects will reorientate until all orientations in the crystal are equally occupied. Below this temperature (10,9 K for H-centers in KCl) defects can be rotated only via their excited state, that means by illumination with light of the wavelength of the absorption band of this center. If this light is polarized only defects with their transition dipole moment not perpendicular to the

electric vector of the incident light can be rotated. After excitation the orientation of the defects will be statistically distributed over the 6 possible orientations. Thus the orientation of the dipoles which were excited will be depopulated, and it is possible to bring defects from one orientation to another.

Rotation of an anisotropic defect involves rotation of the defect induced strain field which causes measurable macroscopic length change of the crystal. Optical absorption measurements with polarized light give information about the defect concentrations in the different orientations.

If this experiment is done in different directions of the crystal the double force tensor describing the elastic interaction of the defect with the lattice can be obtained.

Figure 2 shows the set up for such an experiment. The crystal is mounted inside the cryostat with an
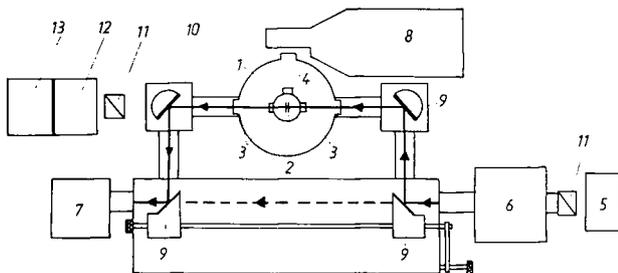

FIG. 2. — Experimental set up. 1) Cryostat. 2) Crystal. 3) Optical window. 4) X-ray window. 5-7) Spectrometer. 8) X-ray tube. 9), 10) Mirror. 11) Polarizer. 12), 13) Light source for bleach.

inductive transducer for length change measurements on top of it. Relative length changes of $\delta(\Delta l/l) = 10^{-6}$ can be measured.

The path of the analyser light is shown by the heavy line. Mirror 10 can be taken off and put in again highly reproducibly in order to do the bleaching with a much stronger light source than the analyser light source. The defects initially are created by irradiation with a 150 kV X-ray tube.

Figure 3 shows the relation between the double force tensor and the induced length change in different directions of the crystal.

In the upper right the 6 different orientations of a $<110>$-defect in the cubic lattice are shown. The tensor describing the elastic dipole can be written in principal defect coordinates or crystal coordinates, according to the directions shown in the upper left.

The length change depends on the strain components in the different directions multiplied by the change of defect concentration in the different directions.

Figure 4 shows a typical absorption spectrum. The solid curve shows the absorption for equally distributed defects. Measurements with $<001>$ and $<010>$ polarized light give the same absorption. Subsequent illumination with light polarized along $<001>$ depopulates all orientations except those perpendicular to $<001>$. Thus the absorption polarized along

Double force tensor in principal coordinates $(\xi.\eta.\zeta)$

$$P = \begin{pmatrix} P_{\xi\xi} & 0 & 0 \\ 0 & P_{\eta\eta} & 0 \\ 0 & 0 & P_{\zeta\zeta} \end{pmatrix}$$

Double force tensor in crystal coordinates $(x.y.z)$

$$P = \begin{pmatrix} P_{xx} & P_{xy} & 0 \\ P_{xy} & P_{xx} & 0 \\ 0 & 0 & P_{zz} \end{pmatrix}$$

Strain components

$$\varepsilon_{xx} = (\frac{a}{2})^{-3} \cdot (S_{11} P_{xx} + S_{12} (P_{xx} + P_{zz}))$$

$$\varepsilon_{zz} = (\frac{a}{2})^{-3} (S_{11} P_{zz} + 2S_{12} P_{xx})$$

$$\varepsilon_{xy} = (\frac{a}{2})^{-3} \frac{S_{44}}{2} P_{xy}$$

Rel. length change

$$\frac{\Delta l}{l}\Big|_{001} = \varepsilon_{xx} \Delta C_{1-4} + \varepsilon_{zz} \Delta C_{56}$$

$$\frac{\Delta l}{l}\Big|_{110} = \frac{1}{2}(\varepsilon_{xx} + \varepsilon_{zz}) \Delta C_{1-4} + \varepsilon_{xx}(\Delta C_5 + \Delta C_6) - \varepsilon_{xy}(\Delta C_5 - \Delta C_6)$$

$$\frac{\Delta l}{l}\Big|_{111} = \frac{1}{3}(2\varepsilon_{xx} + \varepsilon_{zz} - 2\varepsilon_{xy}) \Delta C_{135} + \frac{1}{3}(2\varepsilon_{xx} + \varepsilon_{zz} + 2\varepsilon_{xy})\Delta C_{246}$$

FIG. 3. — Relations between the double force tensor of a defect and the reorientation induced relative length changes in different directions of the crystal.
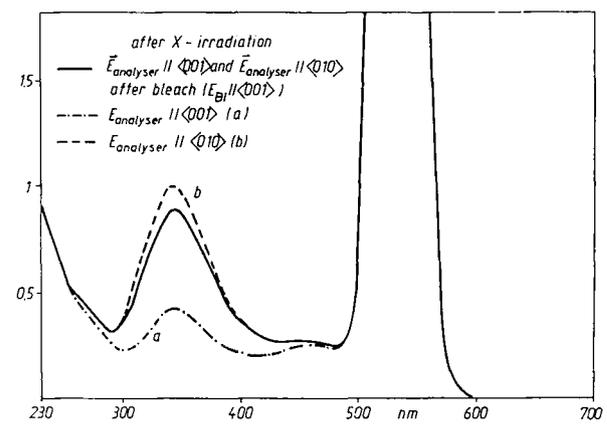

FIG. 4. — Absorption spectrum of KCl after X-irradiation at 8 K.

$<001>$ decreases, whereas the absorption polarized along $<010>$ grows.

Figure 5 shows the results for one direction. The relative length change is measured in $<001>$ direction, and the optical absorption measurement is done for $<001>$ and $<010>$ polarization. The cubes below the drawings show schematically the experimental procedure.

The experiment starts with the irradiated crystal, the defects are equally distributed over to the different
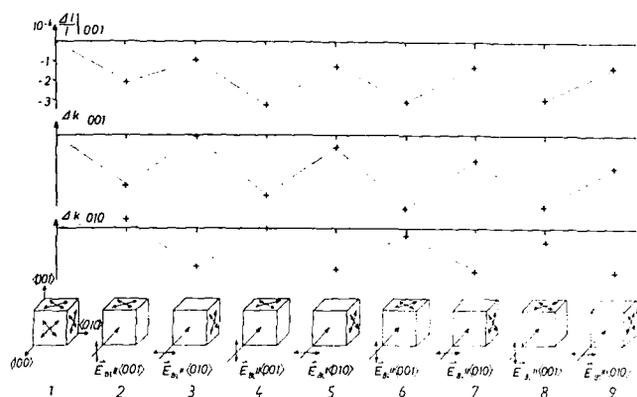
Fig. 5. — Length change, <001>, and <010> polarized optical absorption (345 nm) after bleach of H- and V_K-centers with <001> respectively <010> polarized light.

orientations. Now the crystal is bleached in <100> direction with the light polarized in <001> direction.

The concentration of all defects which are not perpendicular to the electric vector of the bleach light decreases, and the concentration of the defects in orientations perpendicular to the electric vector grows.

After completion of this bleach the concentration in 4 orientations is reduced by approximately 50 %. These orientations are not shown on the schematic anymore, although they are not completely bleached out.

Now the first data points are taken. The total length of the crystal has decreased in <001> direction, that means the defects in the new orientation induce less distortion in this direction. For the next bleach the polarization is turned by 90°, which leads to a reorientation of the defects from the (001) plane to the (010) plane as shown in the schematic. This procedure can be repeated periodically and is shown here for 8 successive steps.

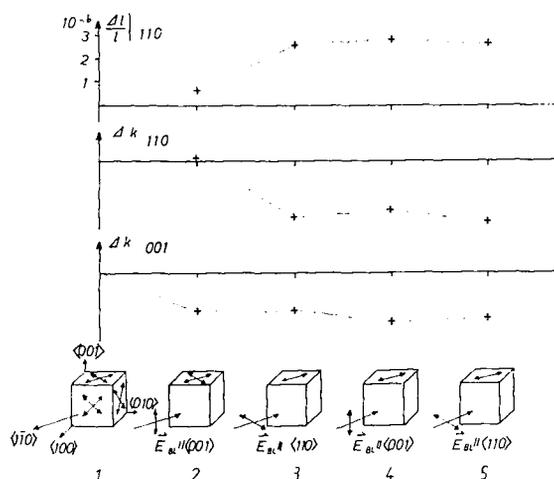Figure 6 shows the results for another measurement.

Fig. 6. — Length change, <001>, and <110> polarized optical absorption (345 nm) after bleach of H- and V_K-centers with <001> respectively <110> polarized light.

The crystal is cut and oriented for relative length change measurements in <110> direction. Bleaching and optical absorption measurements are done in <110> direction.

The schematic at the bottom again demonstrates the procedure. The experiment starts with an equal distribution of the anisotropic defects over the six possible orientations. Bleaching with <001> polarized light in <110> direction depopulates the orientations in plane (100) and (010). Then the crystal is bleached with <110> polarized light. This leads to a saturation for defects with dipole axis along <110> and all other orientations are depopulated. Subsequent change of polarization of the bleach light does not give any change of this stabilized situation. This is verified for the length change measurements as well as for the optical absorption measurements. Only little change can be noticed for the last three sets of data points.

At least 3 sets of measurements in different directions of the crystal are necessary to calculate the double force tensor for the H- and the V_K-center.

Unfortunately it is not possible to do separate measurements for the H- and the V_K-centers, because they are always created together and exist in approximately the same temperature range. It is possible however to create different concentration ratios in different samples and thus separate the double force tensors for the H- and the V_K-center.

The results are :

— Double force tensor for an H-center in KCl

$$p^H = \begin{pmatrix} 6.3 & 0 & 0 \\ 0 & -3.7 & 0 \\ 0 & 0 & 4.3 \end{pmatrix} eV ;$$

— Double force tensor for a V_K-center in KCl

$$p^{V_K} = \begin{pmatrix} 0.6 & 0 & 0 \\ 0 & -0.9 & 0 \\ 0 & 0 & 0.3 \end{pmatrix} eV .$$

Questions : (By LüTY). — As one uses optical reorientation of low symmetry defects for holographic information storage, the interesting question arises from your work, if the length changes from the optical alignment of defects can become large enough (i. e. a sizeable part of one interference fringe), so that it could spoil the read-out process of the volume hologram. Have you thought about this problem, especially in connection with F_A-centers, used for the holograms ?

From our measurements on H- and V_K-centers in KCl we obtained relative length changes due to reorientation in the order of a few parts in 10^{-6}. Although we did not especially study the reorientational behaviour of F_A-centers yet. we made a test measurement on F_A-centers in KCl + NaCl where it turned out that

the length change due to reorientation of F_A-centers is also very small.

(By SPAETH). — What is the minimum concentration necessary of H-centers to determine the double force tensors ? Perhaps you could use the process of photodecomposition of U_2-centers to produce the necessary H-centers without being desturbed by V_K-centers.

The length change due to reorientation should be well above the accuracy which is given by the experimental set up to about $\delta(\Delta l/l) \approx 1 \times 10^{-6}$. A reorientational length change of this minimum detectable size can be obtained with 1×10^{18} cm^{-3} H-centers. So you want to do your measurements at least with about 5×10^{18}-1×10^{19} H-centers/cm^3 to be above the uncertainty of the length measurement.