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OPTICALLY STIMULATED POLARIZATION OF S⁻-VACANCY DIPOLES IN KI

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Résumé. — Le système KI : S⁻ possède trois bandes d'absorption. L'irradiation à - 30 °C cause dans les deux bandes courtes une réaction photochimique. La décomposition des dipôles lacune-S⁻ et la création simultanée de centres F sont observées par des mesures optiques et de thermocourants de dépolariation (ITC).

Lorsque le monocristal dopé est irradié dans la bande d'absorption à 368 nm, on ne trouve pas de réaction photochimique. Quand on applique un champ électrique, cette irradiation augmente la polarisation causée par les dipôles lacune-S⁻ dont l'orientation est déjà gelée dans le réseau cristallin. L'accroissement résultant de la raie ITC est une fonction de la température d'irradiation et est maximale à 179 K — au dessous de la raie ITC (204 K). L'effet est étudié en fonction de la durée d'irradiation et de l'intensité du champ électrique. Un modèle est proposé pour expliquer l'accroissement de la raie ITC.

Abstract. — The KI : S⁻ system has three absorption bands. By irradiation into the two short wavelength bands in the UV, a photochemical reaction occurs. The decomposition of S⁻-vacancy dipoles and the simultaneous formation of F centres is observed by optical and by ITC measurements.

No photochemical reaction is observed when the crystal is irradiated in the near UV absorption band (368 nm). However, this irradiation in the presence of an electric field increases the polarization due to S⁻-vacancy dipoles, the orientation of which is already frozen in the lattice. The resulting increase of the ITC peak is a function of the irradiation temperature. It shows a sharp maximum near 180 K which lies below the ITC maximum (206 K). The effect is also studied as a function of the irradiation time and the strength of the electric field. A model is proposed which explains such an increase in the ITC peak.

Whenever a divalent impurity is introduced in an alkali halide crystal, it is responsible for the creation of the impurity-vacancy dipoles. The dielectric properties of such impurity-vacancy dipoles in divalent anion doped alkali halide crystal have been studied by Kokott [1]. Here we report about some of the results, which we have collected on S⁻ doped KI single crystals through the ionic thermocurrent (ITC) and optical absorption studies.

The normal ITC spectrum of KI : S⁻ system shows its maximum current at 204 K for a heating rate of 0.05 K/s. For this spectrum, the crystal was polarized at 210 K by applying a voltage of 1 500 V for 4 min., and with the electric field still on, the crystal was cooled down to LNT where the electric field was made off. The crystal was then connected to an electrometer and heated at a constant rate up to room temperature for recording the depolarization current. The relevant dielectric relaxation parameters evaluated from this spectrum are : Activation energy for the orientation of the dipoles (E) = 0.61 ± 0.01 eV ; fundamental relaxation time (τ_0) = $(10 \pm 2) \times 10^{-14}$ s.

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Optical absorption measurements [2] on the KI : S⁻ system have shown three characteristic absorption bands which lie at 368 nm, 278 nm and 232 nm. We have observed that at - 30 °C the irradiation in the 278 nm and 232 nm absorption bands is responsible for the photochemical reaction i. e. S⁻ decomposes into S⁻ with the simultaneous formation of F-centres. No photochemical reaction takes place when the crystal is irradiated at LNT in the 278 nm and 232 nm absorption bands. This photochemical reaction has been studied through the optical absorption and ionic thermocurrent measurements. The decrease in S⁻-centre concentration after photochemical reaction is found to be 53 % through the optical absorption studies which is in fairly good agreement with that of 50 % obtained through the ITC measurements. Contrary to this, no photochemical reaction takes place when the crystal is irradiated in the 368 nm absorption band between room temperature and LNT.

However, by irradiation in the presence of an electric field in the 368 nm absorption band below the ITC peak temperature (204 K) more dipoles can be aligned along the electric field direction and consequently the peak of the ITC spectrum increases. This additional *photo-induced polarization* is a function of the

irradiation temperature, irradiation time, polarization temperature and the strength of the electric field. Such an increase of the ITC peak as a function of the irradiation temperature is shown in figure 1. For

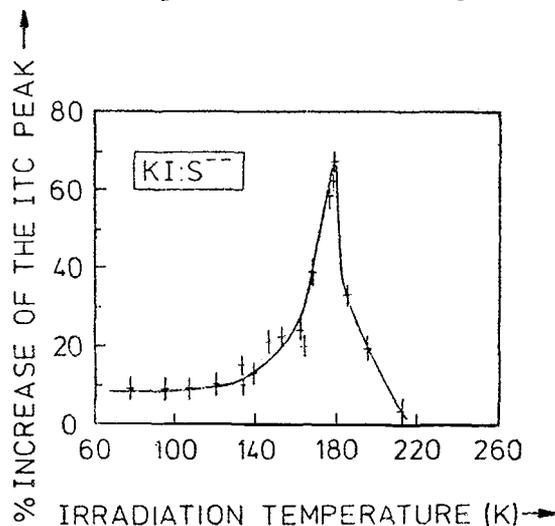


FIG. 1. — Increase of the ITC peak by irradiation in the presence of an electric field in the 368 nm absorption band at different irradiation temperatures; (Polarization temperature = 210 K; Polarizing voltage = 1 500 V; Voltage at the irradiation temperature = 1 500 V. Irradiation time = 210 s).

these experiments, the crystal was polarized at 210 K by applying a polarizing voltage of 1 500 V for 4 min,

and with the electric field still on, the crystal was cooled down to the irradiation temperature where the crystal was irradiated in the presence of the same electric field in the 368 nm absorption band for 210 s. Soon after the irradiation the crystal was further cooled down to LNT where the electric field was made off and the ITC spectrum was recorded. It is apparent from figure 1 that the increase of the ITC peak is a function of the irradiation temperature and has a maximum value at 179 K. It is interesting to notice that even at LNT there is some finite increase which is constant at lower temperatures. The details about the dependence on irradiation temperature, polarization temperature, irradiation time and electric field strength will be published elsewhere [3].

We remark here also on another experiment which can be named as *photo-induced depolarization*. In this case, after the crystal has been polarized, we irradiate into the 368 nm absorption band in the absence of an electric field at a temperature where the polarized dipoles are frozen in. We observe a decrease of the ITC peak.

It can be summarized that by irradiation in the 368 nm absorption band in the presence of an electric field more dipoles can be aligned along the electric field direction whereas such an irradiation in the absence of an electric field reduces the frozen in polarization.

References

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DISCUSSION

R. CAPELLETTI. — Your results can be understood in terms of reorientation of dipoles in the electronic excited state in the same line we proposed for I. V. dipoles in KCl : Pb (R. Capelletti, R. Fieschi, E. Okuno, *Int. Conf. Color Centers in Ionic Crystals*, Sendai (Japan), pag. G. 136 (1974) and for Suzuki phase in KCl : Pb (R. Capelletti, F. Fermi, F. Leoni, E. Okuno, *Proc. of Internat. Symp. on Electrets and Dielectrics*,

São Carlos, Brasil, sept. 1975 (in press). Did you measure the radiative lifetime of the S^{--} -dipole, in order to account for the reorientation? Did you measure the activation energy for the reorientation of the dipole in the electronic excited state? How can you explain the relatively sharp decrease of the photostimulated orientation yield in the high temperature side shown by the slide?