EMISSION SPECTRA OF EXCITONS CREATED IN A SINGLE CRYSTAL OF KBR BY IRRADIATION OF HEAVY IONS AT 4.2 K

Kazuie Kimura and Masashi Imamura

The Institute of Physical and Chemical Research

Although energetic heavy ions are currently in increasing use in solid-state physics and radiation chemistry, experimental information regarding the primary physicochemical processes in irradiated matter is still few ; several theories published so far are not so elaborate for predicting actual processes, either. measurements of the emission spectra from the electronic excited states in the irradiated matter may be one of the useful experimental approaches. A KBr crystal was chosen in the present study because spectral assignments for the excitons and color centers formed in alkali halides by X-, electron-, and photo-irradiation have been well established.

He-, C-, and N-ions of 8 MeV per nucleon from the cyclotron were used and, for pulse irradiation, they were chopped into 40 μs pulses with a repetition of 416 s^{-1}. The beam current was mostly between o.1 and 1 nA, and between o.1 and loo nA for experiments of dose rate effects. Emission spectra were measured under (1) stationary irradiation and under (2) pulse irradiation followed by strobe measurements toobtain the time-resolved spectra. A single crystal of KBr (10 x 10 x 1 mm) was colled at 4.2 K.(Fig.1)

Emission spectra with peaks at 280 and 512 nm were observed for KBr irradiated with C-ions at 4.2 K. They are assignable to the emission due to σ- and π-excitons which are self-trapped excitons known in X- and electron-irradiated KBr. The intensity ratio of the σ- to π-emission (I_σ / I_π) was found tobe 10, which is five times higher than that (2) reported in electron-irradiation. Since it can be verified experimentally that one need not take account of both self-absorption of emission and effect of temperature increase of a sample during irradiation, the possibilties responsible for the higher value of I_σ / I_π are : (1) the larger probability of the radiationless annihilation of the π-excitons, (2) the larger probability of the radiative decay of the σ-excitons, and (3) the preferential formation of the σ-excitons.

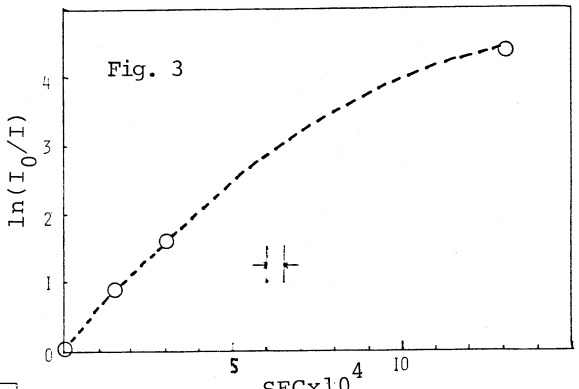
Dense excitationby heavy-ion irradiation may cause interaction between excitons, and between an exciton and a color center, resulting in radiationless decay and in a shorter lifetime of the exciton. The interaction of the π-excitons would increase the ratio, I_σ / I_π . However, following experimetal results indicates that there is no such the interaction. Fig.2 and Fig.3 show time-resolved spectra and the decay curve of the π-emission, respectively. The decay curve can be dissociated into two components, from which a shorter lifetime was calculated to be 0.2 ms. This value is larger rather than that obtained by electron-pulse radiolysis, 0.1 ms [1], and that by laser-photolysis, 20 μ s. [2]. Fig.4 shows that I_σ decreases faster than I_π with increasing doses, in a region higher than 3000 nC. These results indicate that the collisional interaction of π-excitons with π-excitons or a color center hardly occurs. This conclusion is rather natural since π-

exciton is known to be bound tightly at a crystal site. Same discussion can apply in the case of σ-excitons and process (2) should be also ruled out. The process (3) is, therefore, most plausible

Recently, it was reported that free excitons exist for a short time in a photo-irradiated single crystal of KI at 4 K before relaxing into the self-trapped state. In heavy-ion irradiation, the free excitons formed densely in the vicinity of the tracks collide with each other, so that the σ- and π-excitons may be formed in a different ratio from the case of the spontaneous intersystem crossing in X-, electron-, and photo-irradiation.

(1) D.Pooley and W.A.Runciman, J.Phys. C:Solid St.Phys., 3 (1970) 1815

(2) T.Karasawa and M.Hirai, J.Phys.Soc.Japan 40(1976) 755, 769



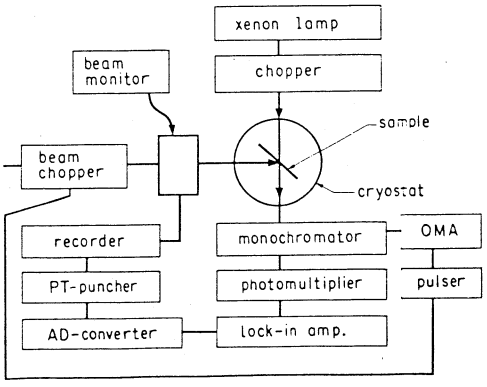


Fig.1