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► **To cite this version:**

A. Nouailhat, G. Guillot, E. Mercier, Truong van Khiem. Polariton effects in the resonant emission of KI. *Journal de Physique Lettres*, Edp sciences, 1978, 39 (13), pp.223-225. 10.1051/jphyslet:019780039013022300 . jpa-00231484

HAL Id: jpa-00231484

<https://hal.archives-ouvertes.fr/jpa-00231484>

Submitted on 1 Jan 1978

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Classification

Physics Abstracts

71.36 — 78.55 — 78.60H

POLARITON EFFECTS IN THE RESONANT EMISSION OF KI

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(Reçu le 5 avril, révisé le 17 mai 1978, accepté le 22 mai 1978)

Résumé. — Sous irradiation électronique, on observe l'émission résonnante analogue à celle obtenue sous excitation optique dans la première bande d'absorption. L'analyse de ses propriétés en fonction de la profondeur de pénétration du faisceau électronique dans le cristal semble montrer un effet de transparence induite par effet polariton.

Abstract. — We have observed resonant emission in a KI crystal under electron irradiation similar to that obtained with U.V. excitation on the first exciton absorption band. Its properties as a function of the penetration depth of electrons in the crystal might be due to a polariton induced transparency.

Measurements on the narrow band edge luminescence in alkali halides are just beginning, but they have already provided important information on the existence of the free exciton and its relaxation processes [1-5]. Nevertheless, free exciton dynamics are not yet well understood; one of the main problems is the polariton propagation mode in the crystal and its interaction with defects. We have undertaken studies on the resonant luminescence of alkali iodides excited by UV light and also by electrons, to be able to vary the depth of the excited layer between about one micron and ten microns. The electron beam is also used to introduce a given concentration of defects in a well defined layer. Nominally pure single crystals of KI and RbI obtained from the Crystal Growth Laboratory of the University of Utah (USA) are cleaved about 1 mm thick and mounted on the cold finger of a helium cryostat in a dry atmosphere. The luminescence is excited in the high energy side of the first excitonic absorption peak with a 200 W deuterium lamp (manufactured by original Hanau) through a grating monochromator (Hilger and Watts D 292), or with a pulsed electron beam, the energy of which is between 10 and 60 kV. The apparatus has been described elsewhere [6]. The luminescence collected from the excited surface of the sample is analysed with a grating monochromator (Mac Pherson 218 A equipped with a 2 400 grooves/mm grating — resolution 13.25 Å/mm —) and detected by a 9 813 QB EMI

photomultiplier followed by a Brookdeal lock-in amplifier.

Experimental results. — Two geometric configurations have been used: electron excitation and observation normal to the sample surface, and observation normal to the excitation, the crystal being at 45° to the excitation as shown in figure 1. The measured emission spectra are the same for both configurations. The maximum energy and width at halfheight of the edge emission near liquid helium temperature are given in table I. It is the same as obtained under UV excitation, and different to that obtained under X-ray excitation (5.79 eV and 5.69 eV respectively for KI and RbI at 5 K [4]). These differences can be attributed to the difference in the excitation mode. X-rays dissipate their energy deep into the crystal contrary to UV or electrons. The resonant emission line is not observed, and only broadening and shifted emission to lower energy is recorded. The behaviours of the emissions are identical considering the temperature under UV and electron excitation in agreement with other results [3, 5]. We have also done a measurement of the resonant luminescence lifetime τ_R of KI at 8 K using an ATNE boxcar (gate width 1 ns): lifetime τ_R has been found to be shorter than that of the σ emission (which represents the limit of our apparatus): $\tau_R < 2.5$ ns.

The properties of the resonant emission for KI at 8 K with the electron irradiation conditions are as follows: the ratio I_R/I_σ , where I_R is the intensity of the resonant emission and I_σ the intensity of the σ component of the

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TABLE I

Excitation	UV		Electrons		X rays [4]	
	E_R (eV)	ΔE (meV)	E_R (eV)	ΔE (meV)	E_R (eV)	ΔE (meV)
KI	5.84 [4]	≤ 14	5.835	≤ 2	5.79	35
RbI	5.734	≤ 1	5.734	≤ 1	5.69	35

E_R : energy of the maximum ; ΔE : width at halfheight.

self trapped exciton emission, is constant if the instantaneous intensity of the current density varies over two orders of magnitude ($0.1-10 \mu\text{A}/\text{cm}^2$ — average energy dissipation rate of $1.3 \times 10^{19} \text{ eV}/\text{cm}^3 \times \text{s}$). I_R is very sensitive to defects : the introduction of $10^{17} \text{ F centres}/\text{cm}^3$ by irradiation at 120 K quenches completely the resonant emission (whereas the intensity of the σ component has decreased by about 10 %). We have measured the ratio I_R/I_σ as a function of the electron energy between 10 and 60 keV, i.e. for a penetration depth x_0 between 1.7 and 19 μ [7] (Fig. 1),

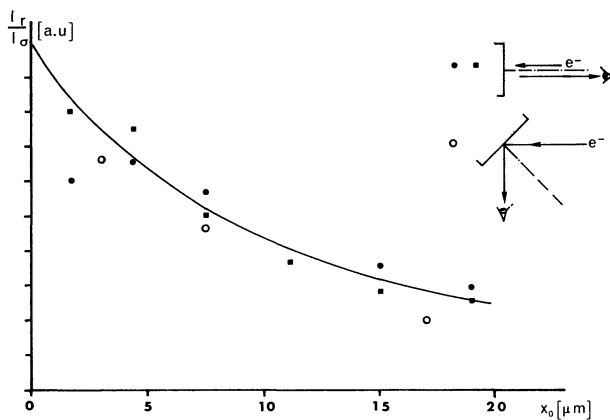


FIG. 1. — I_R/I_σ versus x_0 , $T = 8 \text{ K}$. I_R : resonant emission intensity. I_σ : σ component intensity of the self trapped exciton emission. x_0 : penetration depth of electrons. O, ●, ■ : experimental results.

$$\text{---} : \frac{1 - \exp - u}{u} \text{ with } u = \frac{x_0}{R_0}, R_0 = 5 \mu\text{m}.$$

I_R/I_σ decreases by a factor of about 2 when x_0 increases by a factor of 10. Since the σ emission originates from the whole excited volume, without absorption by the crystal, we can conclude that the resonant luminescence is produced in the volume rather than on the surface of the crystal. Consequently, the absorption for I_R seems anomalously small if we consider that the difference between the peak of the emission E_R and the first exciton absorption peak E_a is about 10 meV [8] : if we suppose that the emission I_R is isotropic [3] in the excited layer, the thickness of which is x_0 and that we can define an absorption coefficient $1/R_0$ for photons of energy E_R , it is easy to show that the ratio I_R/I_σ should vary as $(1 - \exp - u)/u$ where $u = x_0/R_0$. Using our experimental data, we obtain an order of magnitude for $1/R_0 \simeq 10^3 \text{ cm}^{-1}$; on the other hand, the absorption coefficient at E_a measured by conven-

tional absorption is certainly larger than 10^5 cm^{-1} . These results have been obtained with different materials and cannot provide a positive proof for an anomalous behaviour, contrary to the following experiments : defects created by ionizing radiation in alkali halides are known to quench the intrinsic luminescence [9-11]. We have created defects in a 1.7 μm thick layer by irradiation at 120 K, and afterwards we measured the excitation spectrum of the π intrinsic component around the first exciton absorption peak region up to 2 130 \AA , and we observed that UV light is unable to excite the customary π luminescence. This shows that the range for 2 125 \AA photons is smaller than 1.7 μ . Moreover, if we excite the same crystal with 60 kV electrons (over 19 μ range), we cannot detect any resonant luminescence whereas the σ component is only 10 % smaller than in the virgin crystal. The layer of defects inhibits completely the 2 125 \AA emission. This shows that the resonant energy E_R does not behave in the crystal like an ordinary photon.

Discussion. — We deduce from our investigations that the energy E_R corresponding to the resonant emission is able to propagate in the virgin crystal over a large range. We suggest that we are observing a polariton effect, and it might be the first direct evidence for such an effect recently discussed by Fontana [12]

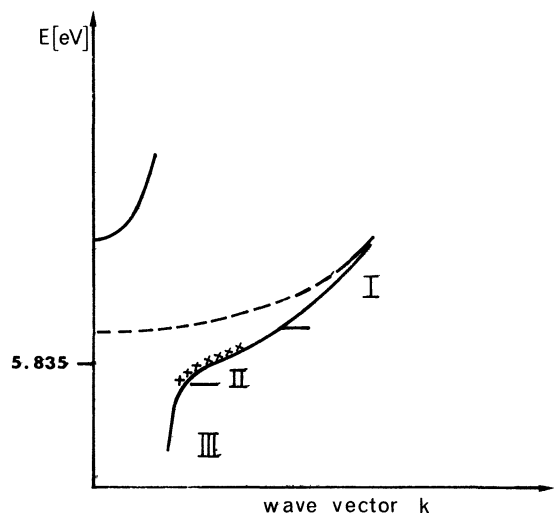


FIG. 2. — Diagram of the dispersion curve of the exciton polariton — — — : uncoupled free exciton (non relaxed exciton) I, II and III are the three energy ranges (see text) xxxxx : approximate location of the bottleneck.

in the alkali halides. Similar conclusions have been made for ZnSe by Sermage *et al.* [13] using a similar method, and for CdS [14], Cu₂O [15] and GaSe [16] from direct absorption measurements, in spite of some criticisms [17].

The excitations of a polar crystal (the excitons and phonons) are strongly coupled to the electromagnetic field [18]. These mixed excitations are called polaritons and can be regarded as *exciton-like* or *photon-like* depending on the considered energy region. The dispersion curve of the polariton consists of two branches, with a peculiar region of anticrossing in which the exciton photon mixing is the largest (Fig. 2). This region of bottleneck (region II, Fig. 2) gives rise to the processes of resonant exciton absorption and emission of light by the crystal [19].

In the process of exciton emission, the polariton wave, which is propagating inside the crystal, reaches the crystal boundary and there changes into a photon which can be detected as the resonance emission.

When defects or impurities are in the path of the polaritons, their energy is dissipated and does not give resonance luminescence which explains its high sensitivity to the defects. In conclusion, it seems that cathodoexcitation is a very good tool to investigate polariton effects in alkali halides directly in the bulk crystal. This avoids all the complications inherent in using the thin films which are necessary for optical absorption spectroscopy, and permits precise measurements of defect-polariton interactions.

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