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THE DIPOLE MOMENT INTERACTION OF BOUND EXCITONS IN HIGHLY EXCITED CdS

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Résumé -- Nous avons trouvé que la polarisation de la luminescence de l'exciton piégé dans CdS dépend de l'intensité d'excitation dans la gamme 10^4 W/cm^2 à 10^6 W/cm^2. Pour interpréter ce phénomène, un modèle d'interaction du moment dipolaire transitoire est proposé. Quand deux excitons sont proches l'un de l'autre, un moment dipolaire induit doit être ajouté à ceux des excitons liés le long de l'axe c. Le moment dipolaire induit croît avec la densité des excitons liés et par conséquent augmente la polarisation de la recombination radiative des excitons liés. Les variations des caractéristiques de luminescence du système uniaxe CdS prédites par ce modèle sont en bon accord avec les résultats expérimentaux.

Abstract -- The polarization of bound exciton luminescence in CdS is found to be dependent on the excitation intensity which varies from 10^4 w/cm² to 10^6 w/cm².To interprete this phenomenon a model of transient dipole moment interaction is suggested. When two excitons approach each other within a very short distance an induced dipole moment should be added to the ordinary ones of bound excitons along the c-axis. The induced dipole moment increases with the density of bound excitons and consequently increases the polarization of the recombination radiation of the bound excitons. The predicted variations of luminescence characteristics of uniaxial CdS from this model agree well with the experimental data.

I - INTRODUCTION

Bound exciton is an impotant localized excited state in crystal.The charcteristies of it's radiative recombination is wide attractive in the fundamental reseach or in the applications.A large number of papers discussing the profile of spectral lines,the energy positions,the intensities of the emission from bound excitons,especially,the detailed structure of the spectra under magnitic fied give us a relatively clear picture about the constitution and the dynamical processes of the bound excitons.//1-7//.In the paper authors propose a new concept for the interaction between bound excitons under high density excitation.In our theoretical model the interaction of the bound excitons on neutral donors is considered as that of neutral atoms.//8//.Because the dielectric tensor has different values at different directions in some uniaxial crystals,the dispersion effect and the induced transient dipole moment are quite different along different directions. In general we choose two directions,which are parallel or perpendicular to the C-axis.For a bound exciton system the energy position and the induced dipole moment can be examined by the polarization of the recombination radiation of bound excitons at different extent of applied excitation.

II - EXPERIMENTAL SETUP AND RESULTS

Very pure and fine CdS single crystal was grown by vapour transfer method*.The

*The authors are much indebted to professor C.Klingshirn in Frankfurt,West Germany for providing this crystal.

as grown crystal was mounted on the holder in cryostat ESR-900 at 77±1K. (see Fig.1)

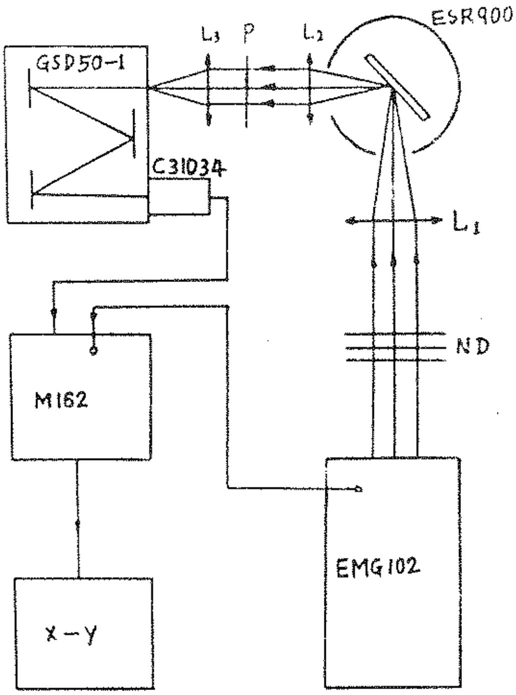

Fig. 1—The experimental setup.ND:the neutral filter,P:the polarizer,L_1,L_2,L_3:focus lenses.

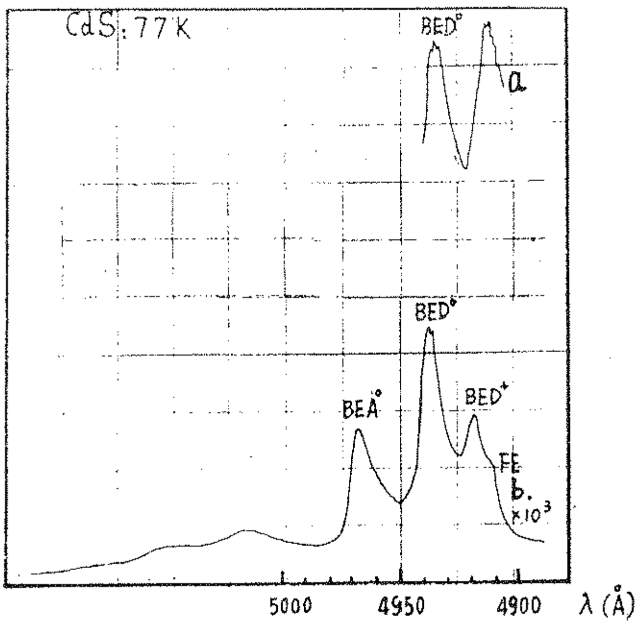

Fig.2 —The photoluminescence spectra of CdS under different I-exc (the brief writting of the excitation intensity). FE:the peak due to the free excitons, BED^+:the peak due to the excitons bound to the ionized donor, BEA°:the peak due to the neutral acceptor.Now pay attention to the peak of BED°. (a).- the spectrum with I-exc of 3×10^4 W/cm²; (b).- the spectrum with I-exc of 1.1×10^6 W/cm². Beside the increase of the luminescence intensity, one can see the longwave shift with the I-excs.

In our experiment a beam of 308 nm light from Excimer laser EMG-102 excited the samples with the electrical field vector E perpendicular to the C-axis of the crystal (i.e E⊥C). Using lenses and neutral filters we can change the I-exc from 10^4 W/cm² to 10^6 W/cm² easily. A polarizer and a double grating spectrometer GSD 50-1 are used to measure the emission peak due to the recombination of BED° in different directions relative to the C-axis of CdS crystal. Through a photomutiplier C-31034 with cooling system the light signal was measured by the M162 boxcar and the X-Y recorder. In Fig.3 we show the shift of the emission peak of BED° and the polarizability P (we'll define the meaning of P in following) of the same one at different excitation intensities.

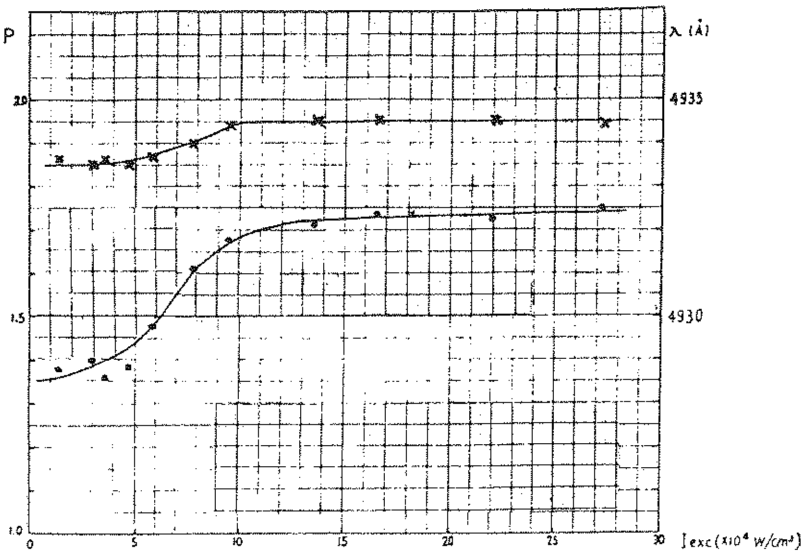

Fig.3—The experimental results of the photoluminescence from BED° in CdS crystal. The cross points stand for the measurement values of λ under different I-excs; the dot points stand for the measurement values of P under different I-excs. The solid lines represent the trial caculation values using the equations (7) and (8) with adequately chosen parameters A,B,and C.

III - Theoresical model

We consider the A sieres Γ_5 exciton only//9//.For simplicity we consider two bound excitons at two neutral donors (BED°) with a distance vector $\vec{r} = \vec{r}_1 - \vec{r}_2$ in an uniaxial crystal, see Fig.4. In our another essay* which aims at the interaction between bound excitons we'll do two things, The first thing is to give the detailed deduction of following equations and the second one is to show that the dispertion effect between BED° and BEA°, between BED° and BED^+ or between BED° and FE are neclegtable and the sing frequency approach is resonable. These two points provide the basic situation for this current paper.

*This work is under mediating

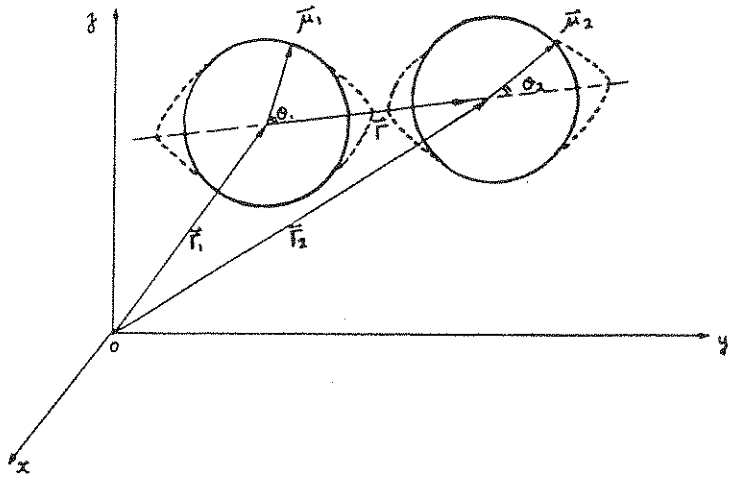

Fig.4—The model of the dispersion action between two nearest BED°. $\vec{\mu}_1$ and $\vec{\mu}_2$ represent the transient electrical-dipole-moment of BED° localized at $\vec{r}_1$ and $\vec{r}_2$, θ_1 and θ_2 represent the angles between $\vec{\mu}_1$,$\vec{\mu}_2$ and $\vec{r}$, $\vec{r}$ is the position vector.

According to the theory of classical electromagnetism, a transient electrical field $\vec{E}_{2-1}$ will be induced at $\vec{r}_1$ by $\vec{\mu}_2$, and the dipole moment $\vec{\mu}_1$ will increase along the vector $\vec{E}_{2-1}$. As the transient emission intensity will be proportional to the square of the transient dipole moment. The latter quantity will be calculated. The square of the added transient dipole moment due to the induction by the nearest BED° can be integrated along the angle θ, the root of the integrated value is calculated and written as M. This quantity M is an increment of dipole moment mainly due to the mutual induction between two nearest neighbours of BED° Then we represent M into M_{11} and M_{1}. i.e paraller and perpendicular to the C-axis. We can obtain following equations:

$$M_{11} = -\frac{\sqrt{2}\,\mu^3}{3r^3K_{11}\varepsilon_o^2\hbar w_o} \qquad (1)$$

$$M_{1} = \frac{\sqrt{2}\,\mu^3}{3r^3K_{11}\varepsilon_o^2\hbar w_o} \qquad (2)$$

$$U = \frac{2\,\mu^4}{3r^6K^2\varepsilon_o^2\hbar^2 w_o^2} \qquad (3)$$

Here, $\mu = \mu_1 = \mu_2$, $\hbar w_o$: the photon energy of the recombination radiation of BED° without the interaction with the neighbour one, K_{11}: the optical inductivity along the C-axis, K_1: that one perpendicular to the C-axis, ε_o: the inductivity in vacuum, U: the energy decrement of the BED° due to the interaction with the nearest neighbours. The detailed deduction of these equations will appear in our another essay, which deal with the interaction between bound excitons. Now we difine the polarizability P as following:

$$P = \frac{I_{11}}{I_1} = D.\frac{(\mu_{11}^{\circ} + M_{11})^2}{(\mu_1^{\circ} + M_1)^2} \qquad (4)$$

Here, I_{11} and I_1: the intensity of the polarized emission of BED° in the direction parallel or perpendicular to the c-axis respectively, D: a proportional constant. Considering $\mu_{11}^{\circ} = \mu_1^{\circ} = \mu^{\circ}$; $M_{11} \gg M_1$ and $\mu_{11}^{\circ} \gg M_{11}$, we obtain:

$$P = D.(1+\frac{\sqrt{2}\,M_{11}}{\mu}) \qquad (5)$$

From equation (3) it shows that, the energy of BED° will decrease due to the mutual interaction, and the emission of BED° will shift to the longwave side with a quantity of $\hbar\Delta w = U$, or

$$\Delta\lambda = \frac{C}{2\pi w_o^2} U \qquad (6)$$

Here C: the light velocity in the vacuum. The deducement of equation (5) is based on the fact that the value of K_{11} is smaller than K_1. If we take account of the optical alignment of excitons in II - IV compounds /7/10//, the value of M_{11} will go up a storey still higher. this fact can be used to understand that the measurement values of P and $\Delta\lambda$ are slight larger than the calculated one by using equations (1) to (3).

Normally the density of BED° in crystal is proportional to the excitation intensity I-exc, so we have the relationship I-exc $\propto r^{-3} \propto M$, the equation (5) and (6) may be expressed as:

$$P = A.(B + \text{I-exc})^2 \qquad (7)$$

$$\lambda = \lambda_o + C.\text{I-exc}^2 \qquad (8)$$

Here, A, B and C are Constants.

IV – The conclusions

1. - the polarizability P of the emission from BED° in CdS increases with the excitation intensity I-exc, and the theoretical relationship between P and I-exc agrees with the experimental results
2. - The photo energy from BED° in CdS decreases with I-exc, and the theoretical relationship between λ and I-exc agrees with the experimental reluts well.
3. - These two phenomena appear in the same similarity. When the I-exc reaches about 10^5 W/cm², both processes will be saturated, because of the saturation of the density, which will be limited by the density of the defects which bound exciton as BED°. From its point of view we can evaluate the density of neutral donor, which is estimated as 10^{18}/cm³.

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