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SPATIAL EXPANSION OF HOT ELECTRON-HOLE PLASMA AT HIGH DENSITY IN CdSe

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Résumé : Nous présentons la première étude expérimentale du transitoire à l'échelle picoseconde de l'expansion d'un plasma chaud et très dense dans CdSe (en plaquette) fortement excité par une impulsion d'un laser YAG (Nd³⁺) à modes synchronisés de durée 30 ps. Nous montrons qu'au-dessus d'une densité critique, le plasma explose à une vitesse qui est supérieure de deux ordres de grandeur à la valeur de la vitesse de diffusion à l'équilibre thermodynamique. Nous utilisons nos calculs théoriques antérieurs pour analyser les résultats.

Abstract : We report here the first experimental study of time resolved expansion in the picosecond time scale of hot electron-hole plasma at very high density generated in CdSe platelets strongly excited by a mode locked Yag laser pulse of duration 30 ps. We show that above a critical density, the plasma expands at a velocity which is two orders of magnitude greater than the thermodynamic equilibrium diffusion velocity value. We use our previous theoretical work for the analysis of the results.

Introduction.- In direct gap semiconductors the electron hole plasma luminescence can be used as a probe for testing the presence of the plasma in a defined region of a sample ; we apply here this idea to the high density plasma in CdSe at low temperature, using appropriate experimental conditions.

1. Experimental set up.- A complete description of our experimental apparatus has been previously reported [1], [2], [3]. In the present experiments, a picosecond pulse (30 ps) is used to excite the sample (Fig. 1). The intensity of the pulse, measured by a photodiode, is introduced into a two levels discriminator which defines a narrow window for pulse energy. When the energy of the excitation pulse falls within this energy window, the two levels discriminator authorizes the processing of the experimental data by multichannel computer. Two photons absorption at $1.064\text{ }\mu\text{m}$ gives rise to homogeneous excitation in CdSe platelets, according to recent measurements [4]. The luminescence of the sample is focalised into a Kerr cell which transmits the incident light only when it is excited by the pulse at $1.064\text{ }\mu\text{m}$, that means during 30ps, and so are obtained the time resolved spectra. The transmitted luminescence enters a monochromator and is finally detected by an optical multichannel analyser monitored by the energy window discriminator of the excitation.

The sample is a platelet, grown in vapor phase, of typical dimensions $1 \times 1 \times 0.03\text{mm}^3$. We have chosen a thin sample for two reasons :

- first, we get an homogeneous excitation ; the inhomogeneities in the excitation are less than 10 % between the front surface and back surface of the platelet ;

- Secondly, the stimulation is low in the detector direction (Y) (see Fig. 2) and we shall neglect it in our calculations.

Fig. 1

Fig. 2

The excited region is a parallelepipedic volume ($1 \times 0.3 \times 0.03 \text{ mm}^3$) (see Fig. 2). The luminescence emerges through a slit (width $100 \mu\text{m}$) in a black paper which covers all the back surface of the sample, but, only are detected the photons which spring in a solid angle of $\pi/100$ steradian, in order to increase the spatial definition of the luminescent plasma. The principle of the experiment is to get a time and wavelength resolution of the luminescence which springs from the back side slit for various positions X of the excited region (Fig. 2) so that we obtain the space and time resolved luminescence spectrum of the sample. The CdSe platelet is stuck on a copper cold finger in an optical cryostat. When the incident power is in the range $100 \text{ MW/cm}^2 < P < 1.5 \text{ GW/cm}^2$, the plasma density is given by $n \approx \beta P^2 (1 + \beta P e)^{-1} (2 h \nu)^{-1}$ (1) where β is the two photons absorption coefficient; $\beta \approx 0.03 \text{ cm/MW}$ for $\lambda = 1.064 \mu\text{m}$ [4], e is the sample thickness, and $h \nu$ is the excitation photon energy. In this work, the plasma density is always calculated using the formula (1); when the incident power increases, the exciton line (present at low excitation) disappears at a density $n = 1.5 \times 10^{17} \text{ cm}^{-3}$, derived in this way, which is in good agreement with previous experimental determination for the Mott density [5].

When the incident power P is lower than 1.5 GW/cm^2 , the lattice heating due to laser pulse is always lower than 15 K [3]. The experiments are performed successively at $T_L \approx (10 - 20 \text{ K})$, $T_L \approx (80 - 90 \text{ K})$ and $T_L \approx 300 \text{ K}$.

2. Experimental results and analysis. - A. Experimental results. - The figure 3 shows two time resolved luminescence spectra due to recombination of electron-hole pairs in the plasma, assisted by the emission of an LO phonon. In the range $300 \text{ MW/cm}^2 < P < 1.5 \text{ GW/cm}^2$, the band gap reduction induces a shift of the luminescence

Fig. 3 : Time resolved L0-phonon assisted plasma luminescence originating from a non-laser excited region ($X = 275 \mu\text{m}$).

but the process is always the same. In a detailed analysis [2] we have shown that at high plasma temperature, the low energy edge is the renormalized band gap E'_G minus the longitudinal optic phonon energy $\hbar\omega_{LO}$ and the upper edge is approximately the renormalized band gap itself. Consequently, this luminescence is an evidence for the presence (at the point X) of high density electron hole plasma. The time resolved spatial distribution of the detected luminescence is strongly dependent on the excitation : under a threshold P_e , the light springs only from the initially excited region, while above this critical value P_e , the light emerges from the whole sample. Fig. 4 shows the luminescence Φ_{λ_0} at a given wavelength in the plasma band versus X for an incident-time averaged-power $P = 1 \text{ GW}/\text{cm}^2$ at two times $t_1 = 50 \text{ ps}$ and $t_2 = 80 \text{ ps}$. The time origin is defined as the luminescence threshold. When P is great-

Fig. 4 : Two post-excitation distributions of time resolved luminescence. - - : Theoretical fit with (8).

Fig. 5 : Time integrated luminescence for two relative directions of propagation with respect to the c-axis (the effect is similar for time resolved luminescence).

ter than a critical value P_c (which is dependent on the relative direction x with respect to the c -axis) the light emerges from the whole sample before the end of the laser pulse. We find $P_c (x/c) \simeq 0.9 \text{ GW/cm}^2$ and $P_c (x \perp c) \simeq 0.8 \text{ GW/cm}^2$.

All these results are similar when the experiment is performed at $T_L \sim 10 \text{ K}$ and $T_L \sim 80 \text{ K}$; on the contrary, we cannot detect any luminescence outside of the excited region when $T_L \sim 300 \text{ K}$.

B. Analysis. - For a quantitative analysis, a diffusion model is used, in which the diffusivity D is only a parameter; the fit of the two curves ($t_1 = 50 \text{ ps}$) and ($t_2 = 80 \text{ ps}$) in Fig. 4 with this model, allows us to give an order of magnitude for D . The spatial plasma density $n(x)$ in the x direction depends on the photon density (luminescence) distribution within the sample via the stimulated emission process [6]; this is a very difficult problem because of the gain saturation of stimulated luminescence. E.O. Göbel and al. [6] proposed a self-consistent model in which $n(x)$ is a symmetric bell-shaped function.

Here, for simplicity, we assume :

$$(2) \quad n(x, t=0) = N \exp - \frac{4 \text{Log} 2}{a^2} x^2$$

where a and N are parameters.

The diffusion is governed by :

$$(3) \quad \frac{\partial n(x, t)}{\partial t} = - \frac{n(x, t)}{\tau} + D \frac{\partial^2 n(x, t)}{\partial x^2}$$

where D and τ are the "ambipolar diffusivity" and the lifetime respectively. The solution of (3) with the initial condition (2) is :

$$(4) \quad n(x, t) = N e^{-t/\tau} (1 + \frac{16 \text{Log} 2}{a^2} x^2 Dt)^{-\frac{1}{2}} \exp - \left\{ x^2 / (a^2/4 \text{Log} 2 + 4Dt) \right\}$$

The luminescence detected in the Y direction (Fig. 2) for a given wavelength λ_0 is a function of x . We call $I_{\lambda_0}(x, t_i)$ the distribution function for spontaneous luminescence intensity at time t_i , with λ_0 given by $\frac{hc}{\lambda_0} - E'_G = \text{constant} (= 11 \text{ meV})$.

For a phonon LO assisted plasma luminescence, we have [2] :

$$(5) \quad I_{\lambda_0}(x, t_i) \propto \int_0^{\hbar\nu + \hbar\omega_{LO} - E'_G} dE_c \sqrt{E_c} \sqrt{\hbar\nu + \hbar\omega_{LO} - E'_G - E_c} |M_{el-ph}|^2 (1 + N_q) f_e(E_c) f_h(\hbar\nu + \hbar\omega_{LO} - E'_G - E_c)$$

where M_{el-ph} is the matrix element for the electron-phonon interaction, E_c is the kinetic energy of the electron, f_e and f_h are the Fermi statistic factors for electrons and holes respectively, N_q is the LO-phonon occupation number and $\hbar\omega_{LO}$ is the LO-phonon energy.

We shall give latter an estimate of the plasma temperature, but in all cases, despite its high density, the hot plasma is rather close a non degenerate state (because the higher the excitation is, the higher the density is, but the higher the plasma temperature is !). If N_q is lower than unity, the formula (5) gives :

$$(6) \quad I_{\lambda_0}(x, t_i) \propto \exp \frac{\mu_e - \mu_h}{kT} \propto n p \simeq n^2$$

where μ_e and μ_h are the chemical potential for electrons and holes respectively, T is the plasma temperature, and k is the Boltzmann constant.

With (6) and (2) we obtain :

$$(7) \quad I_{\lambda_0}(x, t_i) = C [n(x, t_i)]^2 = A(t_i) \exp - \frac{4 \log 2}{b_i^2} x^2$$

where $i = 1$ or 2 and $A(t_i)$ and C are independent of x .

b_i is defined by $b_i^2/4 \log 2 = (d^2/8 \log 2) + 2 D t_i$

Through the slit (Fig. 2), the total measured intensity distribution for λ_0 is :

$$(8) \quad \Phi_{\lambda_0}(X, t_i) = K_1 \int_{X - f/2}^{X + f/2} I_{\lambda_0}(x, t_i) dx = \\ K_2 \left\{ P \left[\frac{\sqrt{8 \log 2}}{b_i} \left(X + \frac{f}{2} \right) \right] - P \left[\frac{\sqrt{8 \log 2}}{b_i} \left(X - \frac{f}{2} \right) \right] \right\}$$

Here K_1 and K_2 are constants, X is the distance between the centre of the excited zone and the centre of the slit (Fig. 2), f is the slit width and $P(z)$ is the tabulated function :

$P(z) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^z \exp -(u^2/2) du$. The best fit of the experimental data (Fig. 4) is obtained for $b_1 = 187 \mu\text{m}$, $b_2 = 230 \mu\text{m}$ at t_1 and t_2 respectively and $\tau \simeq 160$ ps. We deduce $D \simeq 10^6 \text{ cm}^2/\text{s}$. This value of 160 ps for the parameter τ cannot be associated with the spontaneous luminescence only, because the "diffusion" along x is affected by the stimulation of light in this direction. The value for D is more than four order of magnitude greater than the conventionnal diffusivity : the electron hole plasma blows up !

3. Discussion. - The mean kinetic energy of the photoexcited electron-hole pairs is here $(2 h\nu - E'_G) \simeq 0.5 \text{ eV}$. When the density is high enough to permit the internal thermalisation of the plasma, i.e. $n \gg 10^{17} \text{ cm}^{-3}$ [2], a temperature can be defined ; we have calculated the temperature kinetics, but also the time dependent non-equilibrium distributions for the mixed plasmon-LO phonon modes [7] and for TO phonons modes.

Four consequences follow from the use of this analysis for our experimental results :

- Firstly, we can verify the previous affirmation of non degenerate plasma statistics in the first 80 ps (plasma temperature is always greater than 70 K).
- Secondly, the knowledge of the plasma temperature allows us to estimate the thermodynamic equilibrium diffusivity, using the Einstein relation. The calculation gives a value for D_0 at least four orders of magnitude lower than our experimental values of D ;
- Thirdly, we can calculate the absorption coefficient (which is a function of the plasma temperature) and see that the photon transfert [8] cannot be considered as the main mechanism in this rapid displacement of the luminescent plasma. Otherwise, the absence of hot luminescence in the experimental spectra supports this interpretation ;
- Fourthly, the part played by the non-equilibrium distributions of phonons is very important in the cooling of high density electron hole plasma. Moreover, at low lattice temperature, the plasma expands out of the initially excited region in a lattice with few phonons (except the ones created by plasma cooling) whereas at high lattice temperature ($T_L \sim 300 \text{ K}$) it does not expand.

M. Combescot [9] calculated the velocity for the expansion of a very dense plasma at

$T = 0$ K, using an hydrodynamic model ; if we use this model here, we obtain a value which is the tenth of our experimentally deduced result $V = \sqrt{\frac{P}{\epsilon}} \simeq 0.8 \cdot 10^8$ cm/s.

R. Zimmermann and M. Rösler calculated the free energy per pair F in CdS at different temperatures as a function of density [10]. We can calculate the pressure in the plasma using $p = n \left(-\frac{\partial F}{\partial \log n} \right)_{T,N}$, where N is the total number of electron-hole pairs ; when the density is increased above the electron-hole liquid density ($n_0 \sim 5.4 \times 10^{17}$ cm⁻³ in CdSe) this pressure rises up rapidly. This fact is in good agreement with a plasma expansion.

Conclusion. - We have studied the spatial expansion of the luminescent electron hole plasma at high density in CdSe, using a time, space and wavelength resolved picosecond photoluminescence experiment. The main result is that above a critical density, the plasma blow up at a velocity at least two orders of magnitude greater than the thermodynamic equilibrium value. A model for plasma explosion at $T \neq 0$ taking into account the non equilibrium distribution of phonons would be very complex ; we think that the progress in this field depends mainly on the future experiments in various materials to approach the space resolved kinetics of the plasma temperature and non equilibrium distributions of phonons.

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