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PICOSECOND TIME OF FLIGHT MEASUREMENTS OF EXCITONIC POLARITONS

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The group velocity of excitonic polaritons has been measured in CuCl and ZnSe using the technique of picosecond time of flight measurement. Experimental results in CuCl are well explained by the dispersion relation of excitonic polariton including spatial dispersion. Those in ZnSe ascertain the existence of the intermediate polariton branch (IP). The group velocity of IP is much faster than the calculated one using the Fishman's model.

I. Introduction

Recent advances in picosecond optical measuring technique have opened up the possibility of time of flight measurements of excitonic polaritons in semiconductors [1]-[3]. By these measurements it is possible to directly determine the group velocity, and hence the dispersion relation, of excitonic polaritons. These measurements give, in addition, definitive information on the additional boundary conditions (ABC's). In the present paper, CuCl and ZnSe have been adopted as materials to be studied, because they represent two types of zinc-blende semiconductors different in the symmetry of the uppermost valence band.

II. Experiments

Flakes of CuCl and ZnSe crystals grown from the vapor phase have smooth as-grown (111) surfaces. The thicknesses of CuCl and ZnSe crystals were calculated by the use of interference spectra. Tunable picosecond light pulses were generated by means of the optical parametric effect. A $LiNbO_3$ parametric oscillator was pumped by the second harmonics of a repetitively mode-locked YAG:Nd^{3+} laser and the second harmonics of the parametric signals were used for measurements. The samples were directly immersed in liquid helium. A face of the sample was hit by light pulses. The pulses travel in the sample and reappear as light pulses from the other face of the sample. Transient response of the transmitted light intensity was analyzed using a CS_2 optical Kerr shutter. Time resolution is about 20 ps which is limited by the temporal pulse width of the laser light.

III. Experimental Results and Discussion

Using the measured time of flight Δt and the thicknesses d of samples, the group velocity v_g of the pulses in the crystals is calculated by

$$v_g = \frac{d}{\Delta t} \quad . \tag{1}$$

In case of CuCl, as the energy of incident light approaches the Z_3 exciton resonant energy (3.2025 eV), Δt becomes longer and v_g approaches the order of 10^{-5} times the velocity of light in vacuum. In spite of zinc-blende structure, CuCl has the two-fold degenerate (Γ_7) uppermost valence band according to the negative spin-orbit interaction. Therefore the dispersion of the lowest (Z_3) excitonic polariton in CuCl is simplest. It consists of only the lower (LP) and upper (UP) branch polaritons. The dispersion and group velocity of polariton are calculated using the following relations.

$$\left(\frac{ck}{\omega}\right)^2 = \varepsilon_\infty \left(\frac{\omega_\ell(k)^2 - \omega^2}{\omega_t(k)^2 - \omega^2}\right) \quad , \tag{2}$$

$$\omega_t(k) = \omega_t + \frac{\hbar k^2}{2M} \quad , \quad \omega_\ell(k) = \omega_\ell + \frac{\hbar k^2}{2M}$$

and

$$v_g = \frac{\partial \omega}{\partial k} \quad , \tag{3}$$

where $\hbar\omega_t$ = 3.2025 eV (resonant energy of Z_3 transverse exciton), $\hbar\omega_\ell$ = 3.2080 eV (resonant energy of Z_3 longitudinal exciton), ε_∞ = 5.0 and M = 2.5 m_0. The measured group velocity and calculated result are shown in Fig.(1). They coincide well with each other. Above $\sim$ 3.203 eV, the group velocity becomes gradually fast. This presents direct evidence for the existence of the spatial dispersion of excitonic polariton. The translational effective mass of exciton can be determined accurately, because the mass is directly connected to the measured group velocity of LP between $\hbar\omega_t$ and $\hbar\omega_\ell$ as follows,

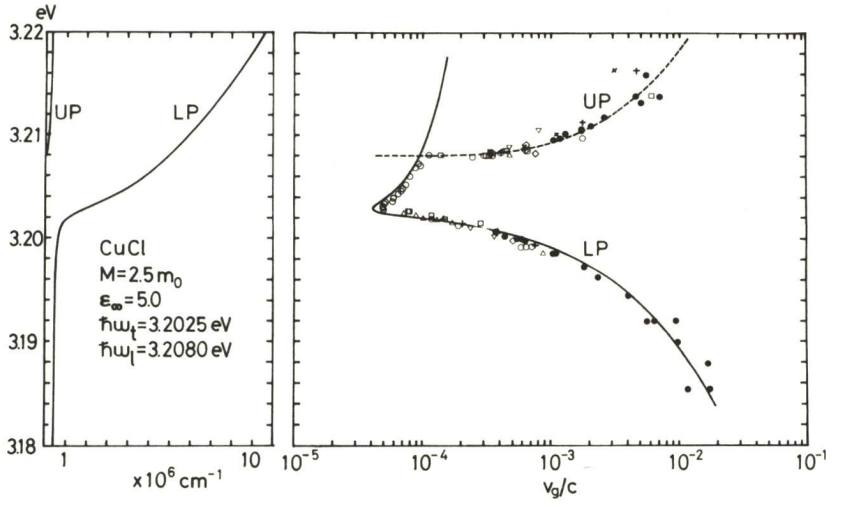

Fig.(1) The dispersion relation of CuCl (left part) and the group velocity of polaritons normalized by c (right part). The dispersion relation is calculated by using eq. (2). Different marks correspond to different samples. The solid and dashed curves illustrate the calculated group velocity

$$v_g \simeq \frac{\hbar k}{M} \quad . \tag{4}$$

The best-fitting procedures lead to the conclusion, $M = (2.0 \pm 0.1)m_0$.

The signal of LP diminishes above $\hbar\omega_\ell$, while that of UP appears instead. These features can be understood as follows. Light pulses above $\hbar\omega_\ell$ are converted to both UP and LP, but the most part of energy flux is given to UP, because UP has much more light-like character than LP. Therefore the share of LP diminishes.

In case of CuCl the uppermost valence band is two-fold degenerate (Γ_7) as mentioned before. On the contrary, the uppermost valence band is four-fold degenerate (Γ_8) in case of ordinary zinc-blende crystals, such as ZnSe, CuBr, CdTe and GaAs. Due to exchange interaction, both the heavy and light excitons couple with each other, so that they are converted to two new normal modes. In addition photon is mixed with these modes. This mixing procedure yields three polariton branches named as upper (UP), intermediate (IP) and lower (LP) branch polaritons as shown in the left part of Fig.(2). The following dispersion relation of excitonic polariton is used for calculation in case of ZnSe [4],[5].

$$\left(\frac{ck}{\omega}\right)^2 = \varepsilon_\infty + \sum_{i=1,2} \frac{4\pi\beta_i(k)\omega_i(k)^2}{\omega_i(k)^2 - \omega^2} \quad , \tag{5}$$

adopting the following parameters : $E_0 = 2.8013$ eV, $E_{LT} = 0.9$ meV, $\Delta = 2.1$ meV, $\varepsilon_\infty = 9$, $M_h = 2.2\ m_0$ and $M_\ell = 0.33\ m_0$, where notations are the same as those in [4] and [5]. These three branches were experimentally confirmed by resonant Brillouin scattering experiments very recently [4]. The present time of flight measurements have also ascertained them. In the right part of Fig.(2) measured group velocities are plotted together with the calculated result using eqs.(5) and (3). As the energy of incident light approaches E_0 from the low

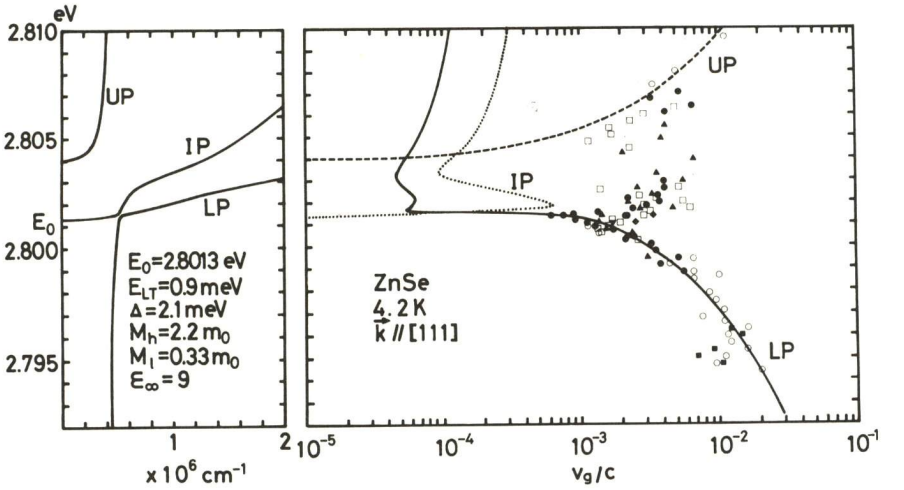

Fig.(2) The dispersion relation of ZnSe (left part) and the group velocity of polaritons normalized by c (right part): The dispersion relation is calculated by using eq. (5). Different marks correspond to different samples. The solid, dashed and dotted curves illustrate the calculated group velocity

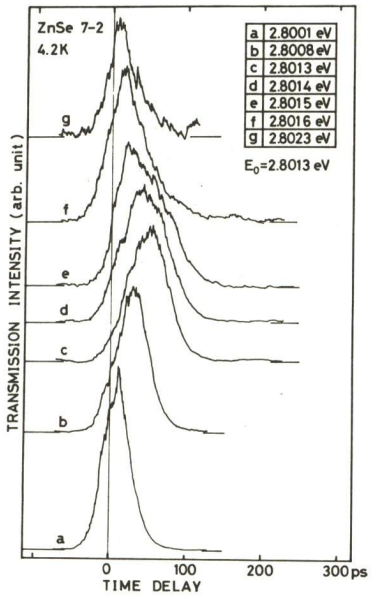

Fig.(3) Transient responses of transmitted light pulses around E_0

energy side, group velocity becomes slower. Around E_0, however, a sudden change was observed as is shown in Fig.(2). Time of flight signals in this energy region are shown in Fig. (3). Comparing these results with calculation, it is concluded that this feature is ascribed to the appearance of the intermediate polariton branch. It should be noted that measured group velocity of the intermediate branch is much faster than calculated one. Agreement is insufficient, even if one changes the value of Δ to 2.1 ± 0.3 meV*. This indicates that the model proposed by Fishman [5] is not satisfactory to describe the dispersion of excitonic polariton in ZnSe. It is also noted that experimental results of resonant Brillouin scattering[4] deviate from calculation above E_0 in the same manner as our experimental results. However discrepancy is largely enhanced in our experiments, because group velocity is the derivative of dispersion $\partial\omega/\partial k$. One possibility to improve the Fishman's model may be to include the k-linear effect.

Above E_0 the share of LP quickly diminishes while that of IP overwhelms. This is because light-like character is larger for IP than LP. The sudden change of time of flight signals around E_0 is due to this competition of LP and IP. At the present stage experimental results are not sufficient for quantitative argument concerning ABC. Conversion efficiencies to LP and IP in ZnSe are not so different from each other as those to LP and UP in CuCl. Hence ZnSe is more promising to this argument of our purpose. In order to obtain direct information to the question which ABC holds, improvement of time resolution is necessary. Using light pulses with temporal width of a few picoseconds, time of flight signals of IP and LP will segregate. In addition this improvement can give more accurate knowledge about the dispersion of excitonic polariton in ZnSe.

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* When one changes the values of parameters within uncertainty ranges described in [4], one finds that v_g of IP is most influenced by Δ in the region a little above E_0.