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Observation of Fabry-Pérot modes in the upper branch of the polariton in ZnSe-GaAs epilayers

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We have studied the reflectance properties of ZnSe epilayers deposited on GaAs substrates by metal-organic vapor-phase epitaxy. For layer thicknesses in the range of a micrometer, reflectance oscillations are observed, and are due to Fabry-Pérot modes in the upper branch of the polariton. This observation is made possible due to the large exciton Rydberg energy in ZnSe and due to the subsequent long-energy extension of this branch of the polariton before its coupling with the $2s$-related exciton branch.

This paper reports the observation of discrete modes in the upper branch of the exciton polariton. The effect has been observed in micrometer-thick ZnSe epilayers grown on GaAs substrates by metal-organic vapor-phase epitaxy. We have studied the evolution of the reflectivity spectra as a function of the thickness of the deposited ZnSe layers. This observation is made possible due to the high crystalline quality of the ZnSe compound and to the intrinsic properties of the compound, such as the large value of the exciton binding energy and the subsequent large energy extension of this branch.

An exciton polariton results from the interaction between an incident photon and an intrinsic exciton. The optically active exciton is coupled to the incident photon via an interaction with the radiation electromagnetic field, on the one hand, and with the excitonic polarization field, on the other hand. Therefore, two coupled propagation modes of light can be defined using Maxwell equations and exciton theory: photonic and excitonic modes (see Fig. 1). Pekar[1] and Hopfield[2] have earlier shown, using quantum-mechanics calculations, that the propagation modes of this polariton differ from the photonlike and from the excitonlike propagating modes. This is particularly true in the region of the resonance energy where the coupling is important, given that the eigenstates of the system have strongly mixed photonic and excitonic dispersion relations. Most of the work to date has been devoted to semi-infinite crystals and good agreement has been obtained between theory and experiment, when models of increasing complexity were used to describe the dielectric constant near a resonance energy.[3] Interesting effects are expected for finite-size semiconductors.[4,5] In contrast to the case of bulk semiconductors, we deal with a system having two finite boundaries. Several cases have been considered depending on the relative value of the layer thickness L_z and with regard to the photon wavelength λ and the Bohr radius a_B of the exciton.[6]

(i) When the thickness of the layer is large compared to the wavelength of the photon ($a_B \ll \lambda \ll L_z$), the distribution of the electronic (and excitonic) states remains quasicontinuous, while the photonic branch is influenced. When the wave

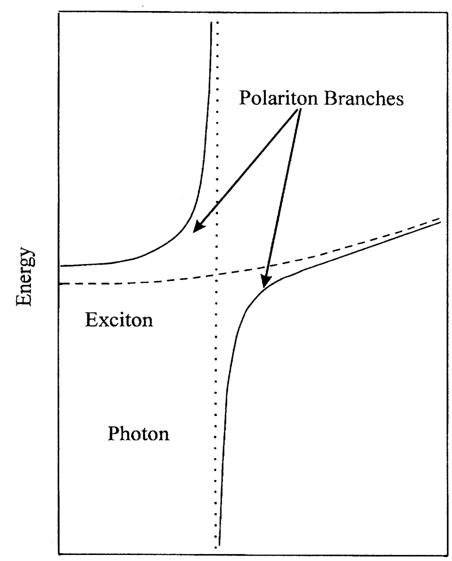

FIG. 1. Schematic diagram of the dispersion relation of the polariton (full line), photon (dotted line), and exciton (dashed line) energy.

function of the upper polariton branch is excitonlike, i.e., for energies $\omega<\omega_L+(\omega_L\omega_{LT})^{1/2}$,[7] quantization occurs in the photon branch polariton following Eq. (1):

$$E(k_\|)=\hbar c\left(\frac{k_\|^2+\dfrac{N^2\pi^2}{L_z^2}}{\varepsilon_b}\right)^{1/2}, \qquad (1)$$

where ε_B is the background dielectric constant, $k_\|$ is the (eventual) in-plane dispersion wave vector. Using a normal incidence condition $k_\|$ is equal to zero.

(ii) Further reducing L_z such that $a_B \ll L_z \approx \lambda$, the quantization can be observed in the excitonlike branch.[8] For the excitonlike branch of the polariton the quantization is given by

$$E(k)=\frac{\hbar^2}{2M}\left(\frac{N^2\pi^2}{L_z}+k_\|^2\right), \qquad (2)$$

where M is the on-axis exciton translation mass. The quantization splitting is now so large in the photonic branch that it cannot be detected. It is important to note that for typical samples where $L_z=\lambda$ and where the active layer is sandwiched between two Bragg reflectors (such that a microcavity is realized) may give exciton-photon Rabi splitting.[9] In the range of energies $\omega_L<\omega<\omega_L+(\omega_L\omega_{LT})^{1/2}$ (here between typically 2.804 and 2.868 eV),[10] we know that the upper polariton branch is photonlike, but its wave function is excitonlike[7] and Fabry-Pérot modes of the upper polariton branch may appear. This has been observed by Kiselev, Razbirin, and Ultrasev[11] in CdSe, as long-period interferences superimposed to smaller period quantization levels in the low polariton branch. Detailed interpretation of the effects reported also for CuCl (Ref. 12) can be found in the review articles of Ivchenko[13] and Bassani and Andreani.[14]

(iii) Last, a third situation can be defined ($a_B \approx L_z \ll \lambda$) which has been studied in detail by Schulteis *et al.*[15] This is at its limit the quantum-well situation.

Figure 2 displays the normal incidence reflectance (top) and the photoreflectance (bottom) spectra of a 0.2-μm-thick ZnSe epilayer. The data have been taken at 2 K and we identify $1s$, $2s$, and $3s$ states of both heavy-hole and light-hole excitons. The light-hole/heavy-hole splitting has been studied in detail by several authors as a function of epilayer thickness.[16] The valence-band splitting increases when the thickness of the epilayer decreases. These studies make the identification of the detected transitions unambiguous. Figure 3(a) displays the reflectance spectra measured at 2 K from a ZnSe epilayer having a nominal thickness of 2 μm. For this thickness the valence-band splitting vanishes and we detect one single bulklike reflectivity structure at 2.8024 eV and $2s$ and $3s$ excited states at 2.8164 and 2.8194 eV, respectively. From these data and from the data shown in Fig. 2, we find the average value of the exciton binding energy in ZnSe to be poorly strain dependent. We note the existence of additional features that are not canceled by photoreflectance spectroscopy which we attribute to Fabry-Pérot modes in the upper branch of the polariton. Figure 3(b) represents the result of a simplified calculation of the polariton dispersion. The dielectric constant is modeled using Eq. (3) with four oscillators[17] (heavy-hole exciton, light-hole exciton, $2s$ and $3s$ levels of the heavy-hole exciton):

$$\varepsilon(k,\omega)=\varepsilon_b+\sum_{i=1}^{4}\frac{4\pi\alpha_{0i}\omega_{0i}^2}{\omega_{0i}^2-\omega^2+B_ik^2-i\omega\Gamma_i}, \qquad (3)$$

where ε_b is the background dielectric constant, ω_{0i} is the

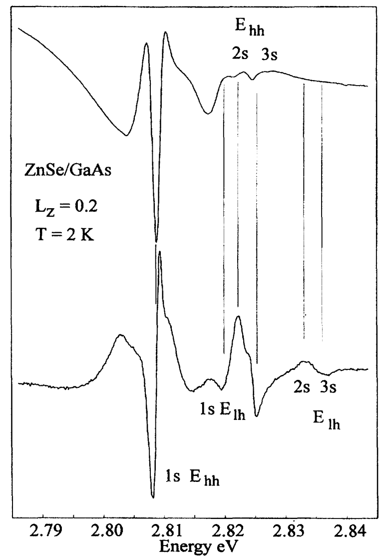

FIG. 2. Reflectivity (top) and photoreflectance spectra (bottom) measured at 2 K from a 0.2-μm-thick ZnSe/GaAs heterojunction. Transitions are observed in the $1s$, $2s$, and $3s$ states for both light-hole and heavy-hole exciton polaritons.

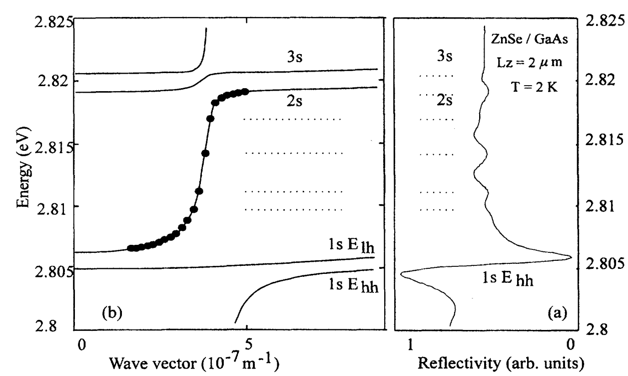

FIG. 3. (a) Reflectance spectra of 2-μm-thick ZnSe layer at 2 K. We note the observation of transitions at intermediate energy between the $1s$, $2s$, and $3s$ exciton polaritons. These oscillations are Fabry-Pérot modes in the upper branch of the polariton. In this sample the valence-band splitting is vanishingly small. (b) Dispersion relation in the excitonic and photonic branches of the polariton (full line). $2s$ and $3s$ light-hole dispersion have not been included in the calculation since the experimental spectra are not detailed enough in the 2.82-eV region. Open circles correspond to quantified values of the wave vector and the corresponding Fabry-Pérot modes in the upper polariton branch.

TABLE I. Relevant parameters used in the calculation, deduced from the line-shape fitting of the reflectivity spectrum.

Transition	$1s\ E_{hh}$	$1s\ E_{lh}$	$2s\ E_{hh}$	$3s\ E_{hh}$
E_{0i} (eV)	2.8024	2.8024	2.8164	2.8188
m_i^* (unit of m_0)	0.89	0.3	0.89	0.89
A_i (eV)	8×10^{-3}	$8\times10^{-3}/3$	$8\times10^{-3}/8$	$8\times10^{-3}/27$
Γ_i (eV)	0.0001	0.0001	0.0001	0.0001

pulsation resonance, Γ_i is the damping parameter, $B_i = \hbar\omega_{0i}/m_i^*$, and m_i^* is the translational mass of the exciton i.

Table I gives the relevant parameters of the calculation obtained after a line-shape fitting of the reflectivity spectrum. We note the excellent agreement between experiment and theory. Quantified values of the wave vector and corresponding eigenvalues are represented using open circles for fundamental polariton branches. Discrete Fabry-Pérot modes can be resolved in reflectance when the interlevel distance is larger than the damping parameters Γ. The observation of a series of short-period oscillations in the low polariton branch like in Refs. 5 (GaAs), 11 and 12 (bulk CdSe and CuCl) is not reported. Their observation is also not reported for energies above ω_L when their period should be doubled (for the discussion of this see Refs. 11–14). This we attribute to slight fluctuations in the thickness of the epilayer leading to overlap of these eigenstates in the lower polariton excitonic branch. From low to high energy, the photonlike branch of Fig. 3, left-hand side, successively displays quasicontinuous states, discrete states, and quasicontinuous states once more. Observation of discrete states of the photonlike polariton branch in epilayers requires samples of well-adapted design. The upper branch of the fundamental polariton must extend over an energy range large enough to contain several values of the quantified wave vector. The energy splitting between ground-state exciton and one of its higher-energy states should be large enough such that quantization of the energy is possible before the onset of mixing with the higher-state excitonic branch. ZnSe grown on GaAs is a good candidate for fulfilling this criterion since the Rydberg energy equals 18 meV, which gives a useful energy range of some 10 meV for the upper branch of the fundamental polariton. In layers thinner than 2 μm, ZnSe is sufficiently strained due to the lattice mismatch with GaAs and due to the difference between thermal expansion coefficients, that the valence band is split into light-hole and heavy-hole subbands.[18] The existence of a light-hole exciton branch between the ground-state heavy-hole exciton and its excited states will bend the photonlike branch more rapidly. Figure 2 illustrates this for a 0.2-μm-thick sample. It is obvious that the presence of the light-hole exciton branch, at 10 meV above the heavy-hole exciton branch in the 0.2-μm-thick sample, reduces the length of the photon branch and explains the inability to observe even one large-period state. The largest valence-

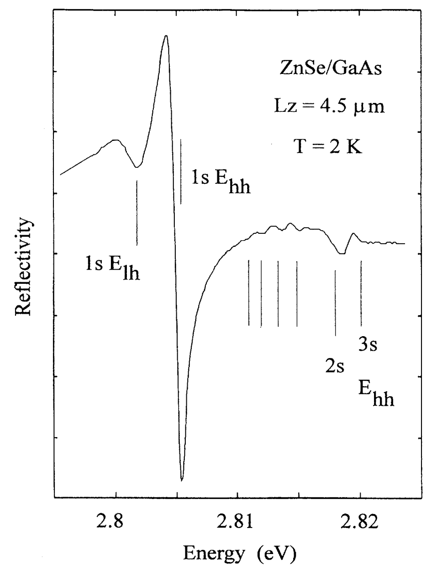

FIG. 4. Reflectance spectrum of a 4.5-μm-thick ZnSe/GaAs epilayer. The residual strain is tensile, thus the lowest state is now the light hole. Discrete levels are observed for high quantum numbers below the $2s$ and $3s$ excitonlike branches.

band splitting is obtained when ZnSe is matched to GaAs, that is, for pseudomorphic layers thinner than 0.1 μm. In that case the resonances become very broad and prohibit the investigation. This broadening is due to the proximity of interface defects and to a lower crystal line quality. Thin (0.2 μm) epilayers represent a hybrid case near the limit of applicability of conditions (i) and (ii). For epilayers of $\geqslant 5$ μm, the experiment becomes *a priori* possible, but the number of quantified levels in the upper branch increases dramatically and the amplitudes of the oscillations are expected to decrease. The reflectance spectrum exhibits the behavior of a semi-infinite semiconductor. In fact, a thickness of 4.5 μm is the limiting value for which we observed Fabry-Pérot modes in the photonic branch of the polariton, as shown in Fig. 4.

In conclusion, Fabry-Pérot modes of the photonlike branch of the exciton polariton are observable in ZnSe layers having thickness ranging between 0.6 and 4.5 μm; this observation is made possible by the exceptionally long length of the photonlike branch of the polariton and is due to the significant value of the Rydberg energy and the exceptional crystalline quality of the samples.[19]

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