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► To cite this version:

A. Tredicucci, Y. Chen, G. Czajkowski, F. Bassani. Excitonic polariton interference in the reflectance of GaAs thin films. *Journal de Physique IV Proceedings*, 1993, 03 (C5), pp.C5-389-C5-392. 10.1051/jp4:1993582 . jpa-00251669

HAL Id: jpa-00251669

<https://hal.science/jpa-00251669>

Submitted on 1 Jan 1993

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Excitonic polariton interference in the reflectance of GaAs thin films

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Abstract: We have investigated the reflectivity of GaAs thin layers in the excitonic absorption region, observing detailed interference structures which reveal the polariton dispersion and the quantization of the exciton centre-of-mass motion. To analyse the experimental spectra, we have calculated the optical properties of thin films using a real-space density-matrix approach adapted to the case of degenerate valence bands. The oscillatory behaviour of the reflectance is well reproduced; a surface layer where the polarization decreases to zero is computed and its thickness is found to be energy dependent. The temperature dependence shows a broadening of the fine structure due to the phonon interaction and a decoupling of excitons and photons at high temperature.

1. INTRODUCTION

In semiconductor thin layers (thickness of a few excitonic Bohr radii), the translational symmetry breaking in the growth direction produces the quantization of the exciton centre-of-mass motion, while it plays a very small role in the relative electron-hole motion.

The phenomenon has been recently studied in GaAs films by observing interference oscillations in the reflectance [1], and emission peaks in the photoluminescence spectra [2].

We here present new reflectivity and photoluminescence measurements on GaAs samples of various thicknesses and with various confining structures at different temperatures [3] and interpret the experimental results with Stahl's real-space density-matrix approach [4-5]. This allows the calculation of the optical properties, without the variational computation of the excitonic wave functions required in the work of D'Andrea and Del Sole [6], and without requiring the introduction of additional boundary conditions [7-8] and of the dead-layer concept [9]. A model electron-hole interaction potential is used together with a phenomenological damping parameter [10-11] and good agreement between theory and experiment is obtained.

In section 2 we give the theoretical details of the calculations. In section 3 we present and discuss the results, comparing the measured spectra with the calculated ones. Section 4 contains concluding remarks.

2. THEORY

We consider a model semiconductor with one conduction band separated by an energy gap E_g from the top valence band degenerate at the Γ point. We also adopt the effective mass approximation

with the cylindrical symmetry for the mass tensor of the valence band. The fundamental equations of Stahl's approach for the coherent wave amplitudes $Y_{H,L}$ are then written:

$$\begin{aligned} \partial_t Y_{H,L} + \frac{i}{\hbar} \left[E_g - \frac{\hbar^2}{2m_e} \partial_{z_e}^2 - \frac{\hbar^2}{2m_{h,H,L}} \partial_{z_h}^2 - \frac{\hbar^2}{2\mu_{H,L}} (\partial_\rho^2 + \rho^{-1} \partial_\rho) + \right. \\ \left. - \frac{\hbar^2}{2M_{H,L}} (\partial_{R_\parallel}^2 + R_\parallel^{-1} \partial_{R_\parallel}) + V_{eh,H,L}(z_e - z_h, \rho) + \right. \\ \left. + V_e(z_e) + V_h(z_h) - i\Gamma_{H,L} \right] Y_{H,L} = \frac{i}{\hbar} M_{H,L} \mathbf{E}, \end{aligned} \quad (1)$$

where H (L) refers to the heavy-hole (light-hole) exciton, z is the growth direction, V_e and V_h are the confining potentials for the electron and the hole, V_{eh} is the electron-hole interaction potential, $\mathbf{E}$ is the electric field, $M_{H,L}$ are the interband transition dipole moments per unit volume and $\Gamma_{H,L}$ are phenomenological damping coefficients.

By introducing appropriate Green's functions $\chi_{H,L}$ [10], these equations can be transformed into integral equations for the polarization $\mathbf{P}$, which is related to the coherent amplitudes $Y_{H,L}$ by the following expression [5]:

$$\mathbf{P}(\mathbf{R}) = 2\Re \int d\mathbf{r} [\mathbf{M}_H(\mathbf{r}) Y_H(\mathbf{R}, \mathbf{r}) + \mathbf{M}_L(\mathbf{r}) Y_L(\mathbf{R}, \mathbf{r})]. \quad (2)$$

Adopting an electron-hole interaction V_{eh} in delta-like form [10-11], we obtain the following material equation:

$$\begin{aligned} \mathbf{P}(z) = \mathbf{P}_H(z) + \mathbf{P}_L(z) = \int_0^L dz' \chi_H(z, z') [2\pi R^* a_0^3 \mathbf{P}_H(z') + M_{0H}^2 \mathbf{E}(z')] + \\ + \int_0^L dz' \chi_L(z, z') [2\pi R^* a_0^3 \mathbf{P}_L(z') + M_{0L}^2 \mathbf{E}(z')], \end{aligned} \quad (3)$$

which can be solved numerically together with Maxwell's equations. R^* and a_0 are the bulk excitonic binding energy and excitonic Bohr radius respectively. Having computed the field and polarization amplitudes $E(z)$ and $P(z)$ inside the slab, and knowing from the constant value ϵ_1 of the dielectric function those in the barrier, we can calculate the optical properties imposing the usual Maxwell's boundary conditions at each interface.

3. RESULTS AND DISCUSSION

The experimental situation for reflectance measurements on semiconductor thin layers is schematized in figure 1. To achieve the best surface quality, the active layer is embedded between some confining

Figure 1: Typical mesoscopic structure used in the measurement of the reflectance of thin layer semiconductors; L , L_1 , L_2 denote thicknesses and ϵ , ϵ_1 , ϵ_2 dielectric constants of the various materials.

structures commonly constituted by $\text{Ga}_{1-x}\text{Al}_x\text{As}$ alloy or by short period superlattices. The inclusion in the calculation of multiple reflections between the various dielectric layers is fundamental to obtain the correct lineshape of the reflectivity [12].

We report in figure 2 the reflectance of a layered structure containing a 150 nm GaAs slab measured at the liquid helium temperature together with the calculated spectrum. We can observe the very good agreement.

By varying the parameters in the computation one can observe some interesting features. All

Figure 2: Experimental (a) and calculated (b) reflectance spectra of a structure with a 150 nm GaAs layer. The following parameters have been used in the computation: $m_{hzH} = 0.34m_0$, $m_{hzL} = 0.094m_0$, $\mu_{\parallel H} = 0.042m_0$, $\mu_{\parallel L} = 0.05m_0$, $m_e = 0.0665m_0$, with m_0 free electron mass; $\Gamma_H = \Gamma_L = 0.25$ meV, $\epsilon = 12.53$, $\epsilon_1 = 11.725$, $\epsilon_2 = 12.0 + 5.0i$; resonances are put at 1515.0 meV and 1515.073 meV [13] for the heavy- and the light-hole exciton respectively and the longitudinal-transverse splitting is chosen to be 0.08 meV [14].

the main interference structures correspond to the space-quantized states of the heavy-hole exciton which has a greater oscillator strength, but caution must be exerted in their identification, because, due to multiple reflections [12] and to polaritonic effects [3], a resonance can result in a dip as well as in a peak. For this layer thickness only the odd exciton states are optically active [3] and therefore the energy position of the structures should be approximately given by:

$$E = E_{ex} + \frac{\hbar^2 \pi^2 (2n)^2}{2M_{exz} L^{*2}}, \quad n = 1, 2, \dots \quad (4)$$

where E_{ex} is the exciton energy at $k = 0$ and M_{exz} is the total exciton mass along the growth direction; the effective confinement thickness L^* is commonly taken as the nominal thickness of the slab minus two dead-layers of the order of the bulk exciton Bohr radius ($a_B \approx 136\text{\AA}$ in GaAs); in the dead-layers excitons cannot exist because of the influence of the surfaces.

Our detailed treatment allows an improvement over this simplified view. We can use equation (3) to calculate *a priori* the effective thickness L^* from the shape of the theoretical polarization, and equation (4) to infer it from the energy position of the reflectance minima after elimination of the confining materials effect [12]. We can see in figure 3 that L^* shows a strong energy dependence. Rapid variations in the *a priori* computed L^* mark the passage from one space-quantized state to the next; the increasing trend is determined by the polariton dispersion near E_{ex} and by surface effects in the relative electron-hole motion on the high energy side.

Finally, we have performed reflectance measurements at different temperatures, finding that the structures for which polariton effects play a major role are characterized by changes in their relative intensities and in their lineshape when the temperature increases [3]. This behaviour cannot be

Figure 3: Effective layer thickness calculated from the energy of the minima in the heavy-hole exciton reflectance (without confining material) by means of relation (4) (stars), and from the theoretical analysis (solid line).

described by a simple increase in the damping factor. We suggest that this is due to a decoupling of the exciton from the photon caused by the phonon field [15].

4. CONCLUSIONS

We have measured the reflectance of GaAs thin layers and have shown that good agreement with theoretical calculations is obtained using a real-space density-matrix approach. The importance of the confining material is revealed and each spectral structure can be explained in terms of polariton interference and quantization of the exciton centre-of-mass motion. The notion of dead-layer is improved and the transition layer thickness is found to be energy dependent. The influence of temperature on polariton features in the reflectance is studied, and evidence of an exciton-photon decoupling is found at high temperatures.

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