

Quantization of excitonic polaritons in thin GaAs layers

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Reflectance, excitation, and luminescence spectra at energies near the 1s excitonic resonance of a thin (500 nm) GaAs-AlGaAs double heterostructure are reported. The reflectance data are analyzed by using a dielectric model. Excitation as well as luminescence spectra reveal the properties of discrete polariton states due to quantization.

I. INTRODUCTION

The excitonic polariton in GaAs has been the subject of extensive studies of the last decade.¹⁻⁷ Most of the absorption, reflectance, excitation, and luminescence experiments on high-quality GaAs crystals grown by vapor-phase epitaxy (VPE) and liquid-phase epitaxy (LPE) have been performed under ambient air conditions. Contamination with oxygen causes charged surface states at the GaAs surface giving rise to an electric surface field. Thus, luminescence and reflectance spectra are strongly affected by the field-induced modifications of the excitonic resonance close to the surface.^{4,5} Cladding of a GaAs layer with two AlGaAs layers prevents contamination with oxygen. In addition, the double heterostructures (DH) grown by molecular-beam epitaxy (MBE) exhibit a well-defined slab geometry with a typical thickness of the central GaAs layer of less than 1 μm .

We report reflectance, excitation, and luminescence spectra of thin high-purity GaAs-AlGaAs DH. The comparison of theoretical and experimental reflectance spectra yields the excitonic eigenenergies and demonstrates a negligible influence of band bending. Reflectance as well as excitation and luminescence spectra are dominated by the spectral features of quantized polariton states due to the small dimensions of the GaAs layer.

II. EXPERIMENTAL

The GaAs-Al_{0.23}Ga_{0.77}As double heterostructure used for our experiments was grown by molecular-beam epitaxy on a (100) oriented GaAs substrate. A thickness of about 500 nm is chosen for the central GaAs layer. The high-purity sample exhibits a residual *p*-type doping level in the 10¹⁴-cm⁻³ range.⁸ During the experiments the specimen is immersed in liquid helium pumped below the λ point. A tungsten iodine lamp is used for the reflectance experiments. Photoluminescence is excited with the 488-nm line of an Ar⁺ laser. Excitation spectroscopy is performed with a tunable cw dye laser system (oxazine 750) pumped by a cw Kr⁺ laser. The reflected or emitted light passes a double grating monochromator with a spectral resolution of 0.015 nm and is detected with a cooled photomultiplier (GaAs cathode). The spectra are recorded with a photon counting system and digital data acquisition. For energy to wavelength conversion we use a factor of 1 239.852 eV nm.⁹ In addition, the refractive index of air is included.¹⁰

III. RESULTS AND DISCUSSION

In Fig. 1 we show the diagram of energy $\hbar\omega$ versus real wave vector \bar{k} , for the excitonic polariton in an infinite GaAs crystal. The coupling of excitons and photons leads to two polariton branches. The upper polariton branch starts at the longitudinal eigenenergy at a wave vector of $\bar{k} = 0$ and becomes photonlike with increasing wave vector. The lower polariton branch is photonlike below the transverse eigenenergy and becomes rapidly excitonlike. In a thin semiconductor layer of thickness *L* excitons as well as polaritons have discrete wave vectors in the direction perpendicular to the layer according to

$$k_{\perp} = \frac{\pi}{L} N, \quad (1)$$

where *N* is a nonzero integer. The eigenstates of this geometry are standing polarization waves (Fabry-Perot modes). Strong coupling between the polaritons inside the semiconductor layer and an external electromagnetic wave is

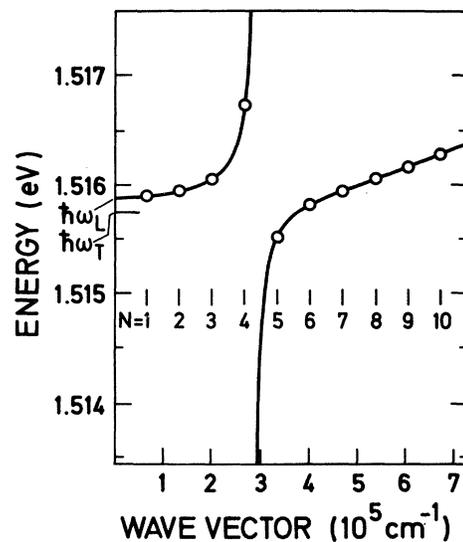


FIG. 1. Energy vs wave-vector dispersion relation of excitonic polaritons in an infinite GaAs crystal. The open circles denote the polariton states with wave vectors with magnitude $k_{\perp} = N\pi/L$, where $L = 470$ nm. The dispersion relation is calculated with an excitonic mass of $0.3m_0$ and longitudinal ($\hbar\omega_L$) and transverse ($\hbar\omega_T$) eigenenergies of 1.515 88 and 1.515 75 eV, respectively.

expected at the energies of the discrete polariton states.

In the upper part of Fig. 2 we show the experimental normal-incidence-reflectance spectra of the GaAs DH. The reflectance structure exhibits three distinct minima and two sharp maxima. The energy position of the reflectance structure is located slightly above the excitonic eigenenergy of bulk GaAs.⁵

Reflectance minima do not suffice to provide information about the excitonic properties. A comprehensive quantitative description of the excitonic polaritons in a semiconductor layer has to include the finite spatial extent of Wannier excitons which causes an exciton-free surface layer, as well as the spatial dispersion which leads to a coupling of the two polariton branches at the boundary. In addition, an electric field caused by charged interface states has to be considered. Microscopic approaches are neither developed for the slab geometry nor are applicable to an inhomogeneous electric field. We therefore describe the resonant behavior of the excitonic polariton in a GaAs layer by a dielectric model neglecting spatial dispersion but taking into account an electric field and an exciton-free transition layer. Details about this model have been reported elsewhere.¹¹ For the refractive index of the AlGaAs layer and the GaAs substrate we take $n_{\text{AlGaAs}} = 3.46$ and $n_{\text{subst.}} = 3.64 + i0.08$ ($i = \sqrt{-1}$).¹²

The calculated reflectance spectrum is included in the upper part of Fig. 2. The agreement between theoretical

and experimental spectra is very good. The fitting procedure directly yields the excitonic eigenenergies and the damping. In addition, we obtain the field parameters of an electric surface field, and we can deduce the actual thickness of the GaAs layer.

The transverse and the longitudinal eigenenergies obtained from the fit are 1.51575 and 1.51588 eV, respectively. The shift of 0.72 meV to higher energies is due to a hydrostatic compressive stress of 190 kp/cm² which is caused by the lattice mismatch at the GaAs-AlGaAs interface.^{1,13}

The line-shape analysis reveals only a negligible influence of an electric surface field. Charged interface states which are responsible for the surface field are mainly discharged by the photogenerated free carriers. This result is substantiated by the missing reflectance spike at the longitudinal eigenenergy in the present study which would be typical for reflectance spectra of GaAs samples exhibiting an electric surface field.^{5,11}

The damping of 0.2 meV obtained from the fit is considerably larger than in bulk GaAs.^{5,11} This result can be explained by a level broadening due to scattering on impurities and/or inhomogeneous strain which both reduce the excitonic lifetime.

Comparison of the reflectance data with Fig. 1 shows that the two sharp reflectance maxima at 1.5155 and 1.516 eV correspond to the $N=5$ and $N=3$ Fabry-Perot modes of the polariton. The slight reflectance maxima above 1.5164 eV may be attributed to the $N=4$ Fabry-Perot mode. The two small reflectance minima above the longitudinal and below the transverse eigenenergies are caused by an interference of the nonresonant excited Fabry-Perot modes of the polariton. In the energy range of the longitudinal-transverse splitting the absorption is strong and these simple qualitative arguments do not hold. Interference of the $N=1,2$ and the $N \geq 6$ Fabry-Perot modes is suppressed. An additional interference structure due to Fabry-Perot modes of the excitonlike polariton branch well above the longitudinal eigenenergy is not observed. This result is contrary to previous reflectance investigations in II-VI semiconductors¹⁴ and can be explained by a higher damping of the polariton in GaAs compared to the small oscillator strength.

The structure of the reflectance spectrum is caused by an interference of all the constituent layers of the DH. Our simple considerations cannot describe *a priori* whether a reflectance minimum or a maximum corresponds to a Fabry-Perot mode. The qualitative attribution of the observed reflectance features to Fabry-Perot modes is supported by the line-shape analysis of the reflectance data which yields a layer thickness of 525 nm. The dispersion relation of the excitonic polariton in Fig. 1 is calculated for an excitonic mass of $m = 0.3m_0$ (Ref. 1) and an effective layer width of 470 nm which includes two exciton-free surface layers of a thickness of the Bohr diameter of 27 nm.¹

The central part of Fig. 2 shows the excitation spectrum obtained from the GaAs DH. Detection was made at the broad emission band at 1.5113 eV. The excitation spectrum exhibits two small emission bands at 1.5128 and 1.5117 eV attributed to "defect-induced" bound excitons.¹⁵ In the vicinity of the excitonic resonance a strong emission peak having a doublet structure is observed. The peak positions of 1.5155 and 1.516 eV exactly coincide with the sharp reflectance maxima. On the high-energy side a weak unresolved interference structure is detected. The observed excitonic emission peak at 1.5155 eV is due to the resonant

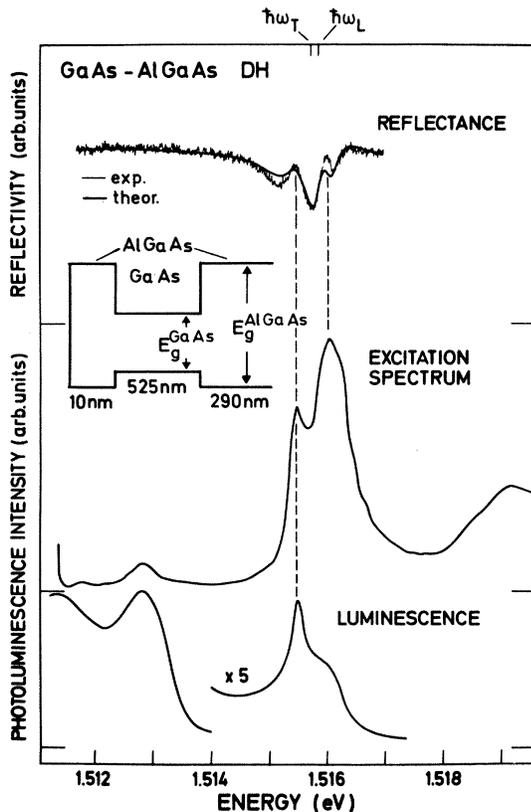


FIG. 2. Normal incidence reflectance, excitation, and luminescence spectra of the GaAs-AlGaAs DH. The experimental reflectance spectrum is compared to a theoretical one. The upper scale gives the longitudinal ($\hbar\omega_L$) and the transverse ($\hbar\omega_T$) eigenenergies. The excitation spectrum is detected at 1.5113 eV. The luminescence spectra is obtained at 0.4 W/cm². The inset shows the schematic real-space energy-band diagram of the DH.

excitation of the $N=5$ Fabry-Perot mode, whereas the stronger emission peak at 1.516 eV is mainly caused by the $N=3$ Fabry-Perot mode. An additional excitation of the discrete polariton states with $N \geq 6$ and $N \leq 2$ may explain the broadening of the high-energy emission peak. From the reflectance data the coupling efficiencies to the Fabry-Perot modes of higher order are expected to be small. The excitation spectrum confirms this result by the observed weak interference structure on the high-energy side of the excitonic emission band.

In the high-energy range at 1.5915 eV the recombination of excited $n=2,3$ excitons and the excitonic continuum states (band gap) is observed. The binding energies of the excited excitons increase because their spatial extents are comparable to the layer width of about 500 nm. Thus, the excitonic eigenenergies of the excited excitonic states decrease. In addition, the finite layer width is also expected to reduce the lifetime of the excited excitons resulting in a smearing of the excitonic emission lines near the continuum threshold towards lower energies.

The excitation spectrum shows directly the absorption related features rather than just a convolution with the competing efficiencies of the recombination channels. Our observed excitation spectrum is in contrast to previous investigations where the excitation spectra are dominated by impurity-related emission.³

In the lower part of Fig. 2, a luminescence spectrum obtained at an excitation intensity of 0.4 W/cm² is shown. The luminescence spectrum in the near-band-gap region exhibits two strong emission bands of the defect-induced bound excitons.¹⁵ At 1.5155 eV a sharp emission line with a small shoulder on the high-energy side originating from free excitons is observed. The energy position of the excitonic emission line exactly coincides with the maxima of the reflectance and excitation spectra at 1.5155 eV.

The refractive index of the GaAs layer in the near vicinity of the $N=5$ Fabry-Perot mode is larger than that of the adjacent AlGaAs. Assuming conservation of the wave vector parallel to the layer k_{\parallel} , only polariton states with $k_{\parallel} \leq k_0$, where k_0 is the wave vector of the emitted light, are not totally reflected at the boundary. Only a small fraction of polariton states radiate into the adjacent medium. The emission of one Fabry-Perot mode is given by a nearly single energy level. Thus, the energy position as well as the half-width of the emission lines are expected to reveal the properties of the discrete polariton states.

The luminescence line at 1.5155 eV can be attributed to the emission of the $N=5$ Fabry-Perot mode. The high-energy shoulder may correspond to the emission of the other discrete polariton states of higher energy.

The luminescence spectra in the near-band-gap region strongly depend on the excitation intensity. In Fig. 3 we depict luminescence spectra obtained at four characteristic excitation intensities. At a low excitation level of 0.4 W/cm² the spectrum is dominated by the recombination of defect-induced bound excitons. With increasing excitation intensity the polariton emission becomes dominant (see for 4 W/cm²). The luminescence peak of the polariton emission remains at a fixed energy position and broadens symmetrically. In addition, an exponential high-energy tail is seen starting 4 meV above the excitonic resonance. At a high excitation level of 28 W/cm² the luminescence line becomes asymmetric because of an overlap with the bound exciton emission line.

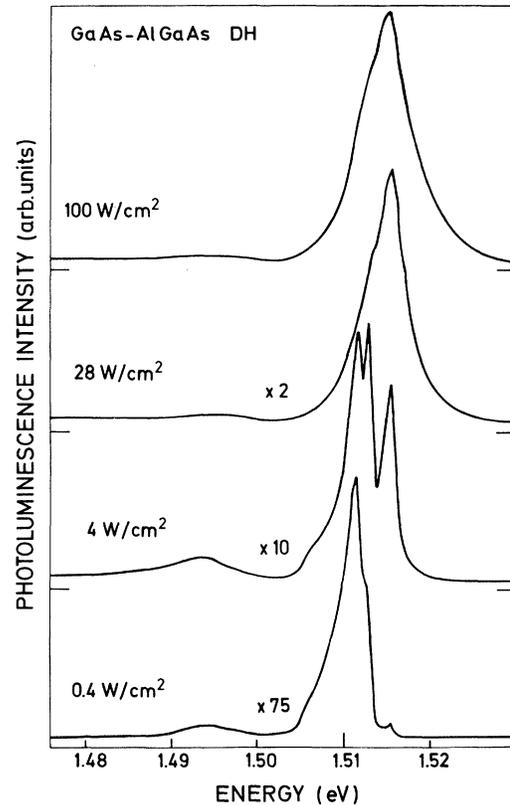


FIG. 3. Luminescence spectra of the GaAs-AlGaAs DH for various excitation intensities.

Our observation of a fixed energy position of the excitonic emission line strongly confirms the discrete nature of the polariton emission. The line shape of the discrete polariton emission lines reveals the damping of the Fabry-Perot modes rather than the mean kinetic energy. A thermal distribution is only reflected by the relative heights of the different polariton emission lines.

The symmetric broadening of the luminescence line with increasing excitation intensity has to be considered by a reduced lifetime due to exciton-exciton and exciton-carrier collisions.

The excitonic continuum states are only slightly affected by the small dimensions of the layer. The energy states are continuous. The emission is thus expected to display the convolution of the density of states and a thermal distribution according to a Fermi-Dirac statistic.¹⁶ This emission behavior is observed by the exponential high-energy tail of the excitonic emission line. The excitonic temperature taken from the slope of the exponential high-energy tail of the excitonic continuum states rises up to 45 K for an excitation intensity of 100 W/cm².

The present luminescence results are in contrast to previous luminescence investigations on LPE- and VPE-grown GaAs crystals,^{1,3,7} where the polariton emission exhibits a doublet structure with a dip exactly at the longitudinal eigenenergy. The two broad emission bands have been attributed to a recombination from upper and lower polariton branch. Because of an overlap with excited donor-bound excitons (from the residual n -type doping) the spectral features of the polariton emission could not be resolved on

the low-energy side. With increasing excitation intensity the high-energy emission band revealed a blue shift.¹

IV. CONCLUSIONS

We have investigated excitonic polaritons in a thin GaAs layer by means of reflectance, excitation, and luminescence experiments. Comparing the experimental reflectance spectra with theoretical ones we obtain the excitonic eigenenergies. Reflectance, excitation, and luminescence spectra re-

veal the spectral features of discrete polariton states due to the finite size of the GaAs layer.

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¹D. D. Sell, S. E. Stokowski, R. Dingle, and J. V. DiLorenzo, *Phys. Rev. B* **7**, 4568 (1973).

²D. L. Hill, *Solid State Commun.* **11**, 1187 (1972).

³U. Heim and P. Hiesinger, *Phys. Status Solidi (b)* **66**, 461 (1974).

⁴B. Fischer and H. J. Stolz, *Appl. Phys. Lett.* **40**, 56 (1982).

⁵L. Schultheis and I. Balslev, *Phys. Rev. B* **28**, 2292 (1983).

⁶C. Weisbuch and R. G. Ulbrich, *Phys. Rev. Lett.* **34**, 654 (1977).

⁷F. Askary and P. Y. Yu, *Solid State Commun.* **47**, 241 (1983).

⁸H. Jung, A. Fischer, and K. Ploog, *Appl. Phys. A* **33**, 9 (1984).

⁹CODATA Bull. **11**, 7 (1973).

¹⁰H. S. Stewart and R. F. Hopfield, in *Applied Optics and Optical En-*

gineering, edited by R. Kingslake (Academic, New York, 1965), Vol. I, p. 127.

¹¹L. Schultheis and J. Lagois, *Phys. Rev. B* **29**, 6784 (1984) (this issue).

¹²H. C. Casey, Jr., D. D. Sell, and M. B. Panish, *Appl. Phys. Lett.* **24**, 63 (1974).

¹³R. Dingle and W. Wiegmann, *J. Appl. Phys.* **46**, 4312 (1975).

¹⁴See, for instance, V. A. Kiselev, *Solid State Commun.* **43**, 471 (1982).

¹⁵H. Künzel and K. Ploog, *Appl. Phys. Lett.* **37**, 416 (1980).

¹⁶H. H. Bebb and E. W. Williams, in *Semiconductors and Semimetals*, edited by R. K. Williardson and A. C. Beer (Academic, New York, 1972), Vol. 8, p. 286ff.