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Induced absorption spectroscopic determination of exciton binding energies in type-II GaAs/AlAs superlattices

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The binding energies of magnetoexcitons in a range of GaAs/AlAs type-II superlattices have been measured directly for the first time, using a novel induced far-infrared absorption technique. Zero-field binding energies in the range 13.2–17.4 meV are found, and these are in excellent agreement with recently published variational calculations. Good fits to the experimental data in the range $B=0$–8 T are obtained with a modified quasi-two-dimensional magnetoexciton theory, allowing exciton dimensionality parameters to be determined.

Advances in the molecular-beam epitaxial growth of short-period GaAs/AlAs heterostructures (with GaAs layers <3.5 nm thick and AlAs layers >1.6 nm thick) have recently resulted in so-called type-II superlattices,[1–3] where the lowest-energy optical transitions are indirect in both real and reciprocal spaces.

This type-II electronic structure results in radiative lifetimes determined by the strength and character of the interface potential mixing Γ- and X-like electronic states and low-temperature lifetime values of a few microseconds are typical.[4,5] Photoluminescence measurements of the indirect transition energies can yield a rather direct measure of the heterojunction band offset values[6] if the binding energy of the type-II excitonic state is known with sufficient accuracy and this provided part of the motivation for the present work.

Here we measure the energy of the transition between $1s$ and $2p$ type-II excitonic states directly by measuring the small changes in the far-infrared (FIR) absorption spectrum of type-II superlattices due to an optically created exciton population. The long lifetimes mean that substantial exciton populations, thermalized to the lattice temperature (4.2 K in all these experiments) can be created, allowing good signal-to-noise photoinduced far-infrared absorption (PFA) spectra to be obtained.

A similar technique has recently been applied to bulk semiconductors.[7] For studies of low-dimensional structures though, the technique has the important advantage that it is "local," i.e., the measured PFA transitions are largely insensitive to the large inhomogeneous linewidths (due to the carrier confinement energy dependence on interface fluctuations) which blur excitonic energy-level structure in interband spectroscopy experiments.

The samples studied (Table I) were all unintentionally doped and grown by molecular-beam epitaxy (MBE) on semi-insulating GaAs substrates. They gave photoluminescence (PL), photoluminescence excitation (PLE), and low-temperature luminescence lifetime data similar to that previously reported.[3,4] The specimens were wedged, cooled to 4.2 K in helium exchange gas, and optically pumped with ≈ 600 mW cm^{-2} of $\lambda=514$-nm laser radiation which was 100% modulated at ≈ 50 kHz. The fractions of the pump radiation absorbed in different regions of the sample (Table I) were calculated from averages of the published bulk absorption coefficients of AlAs (Ref. 8) and GaAs (Ref. 9) and correspond to mean exciton densities of $\approx 1.2\times10^{10}$ cm^{-2} per superlattice period.

FIR radiation from a Beckmann Fourier-transform

TABLE I. Parameters of type-II superlattice structures studied.

Sample	GaAs thickness (nm)	AlAs thickness (nm)	$B=0$ $1s$-$2p$ energy (meV)	70.5 μm peak field (T)	Radiative lifetime (μsec)	Number of periods	% pump light in SL	Exciton binding energy (meV) Experiment	Theory (Ref. 14)	D	$\mu_\parallel^*/\mu_\perp^*$
a	2.54	3.96	11.8	· · ·	6.6	60	70	13.2	≈ 14		
			13.0	· · ·				14.6			
b	2.26	2.26	15.5	4.438	1.0	100	68	17.4	16.8	0.45	0.039
c	2.83	2.26	13.0	4.740	≈ 0.4	100	73	14.6	15.6	0.37	0.058
			14.6	4.369				16.5		0.43	0.044

spectrometer was passed through the pumped sample area and through cooled black polyethylene and wedged TPX filters before being detected with a Ge:Ga extrinsic photoconductive detector (sensitive to 133–40 μm radiation). The PFA signal (with a total strength $\approx 0.05\%$ of the spectrally integrated FIR signal transmitted straight through the sample) was detected with a lock-in amplifier and the resulting PFA spectrum was corrected for instrumental effects by ratioing with the "straight through" FIR spectrum.

Control experiments with the same setup but with 2-μm-thick unintentionally doped MBE GaAs layers ($n \approx 3\times10^{14}$ cm^{-3}) and with type-I quantum-well samples gave PFA signals which were more than 100 times weaker than the type-II superlattice signals. Trials with bulk silicon samples gave similar spectra (with identical peak widths) to those reported in Ref. 7, demonstrating that the sample temperature remained close to 4.2 K.

For the magnetic-field work samples b and c were placed in a calibrated 8-T superconducting solenoid (with the B field parallel to the growth axis) and illuminated with the 70.5- and 118-μm lines from an Edinburgh Instruments methanol FIR laser. The transmitted FIR radiation was waveguide coupled to the detector which was placed outside the solenoid. The effects of the residual B field on the detector and of FIR laser power fluctuations (in total $\approx 50\%$) were again eliminated by electronically taking the ratio of the photoinduced and "straight-through" FIR signals.

In both configurations the strengths of the features in the PFA spectra associated with the type-II superlattice were proportional to the 514-nm pump intensity, up to the maximum achievable level of ≈ 600 mW cm^{-2}.

A typical $B=0$ PFA spectrum (Fig. 1) shows two reproducible peaks at photon energies of 13.0 and 14.6 meV, superimposed on a broad featureless free-carrier absorption background. Two samples (Table I) gave double peaks each with a full width at half maximum width of ≈ 0.6 meV while the third (sample b) gave a single peak ≈ 2.1 meV wide.

The magneto-PFA spectra with 118-μm radiation contained the expected free-carrier cyclotron-resonance (CR) peaks from the GaAs capping and buffer layers, together with strong dips (Fig. 2) at the bulk GaAs $1s$-$2p^+$ shallow donor resonance positions. The latter are due to the optically created hot-carrier population thermally ionizing the residual donor states in the bulk GaAs cladding layers and causing an induced *reduction* in the FIR absorption at the bound donor state transition resonances.

With the 70.5-μm FIR line, samples b and c both showed clear additional features in the 4.3–4.7-T region (Fig. 3). These had the same sign as the CR peak and their integrated peak area was typically $\approx \frac{1}{50}$ of the $1s$-$2p^+$ feature (at ≈ 7.7 T) which was the only other detectable feature at this FIR wavelength.

The peaks in the $B=0$ spectra (although too energetic to be attributed to any known GaAs donor transitions) approximately coincide with the 15.18-meV $1s$-$2p$ acceptor transition measured in photothermal-ionization-spectroscopy (PTIS) studies of carbon-doped GaAs (Ref. 10). The appearance of two lines in our samples, however, combined with the sample-to-sample variation in their energies and their absence in the bulk GaAs epilayer control experiments argues strongly against this assignment. Further evidence comes from the qualitative correlation between the $B=0$ peaks in samples b and c and the respective 70.5-μm magneto-PFA spectra; the carbon $1s$-$2p^+$ line only moves slowly with field,[10] reaching ≈ 15.49 meV by 8 T and would thus not appear on the 70.5-μm/17.56-meV magneto-PFA spectrum.

The peaks in the 4.2–4.7-T range in the 70.5-μm magneto-PFA spectrum do not appear in any published

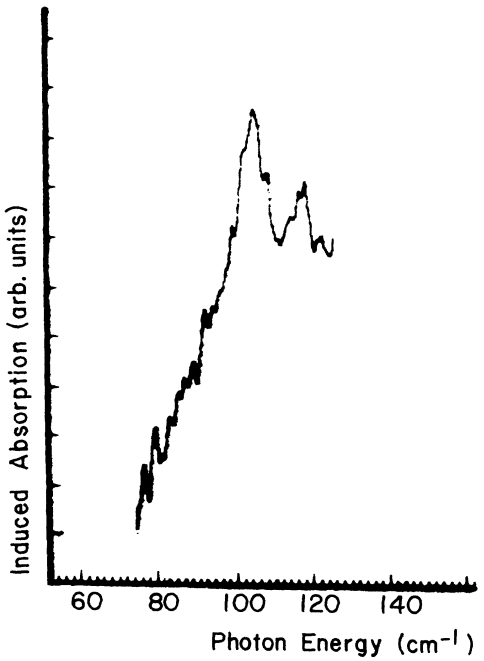

FIG. 1. Normalized FIR photoinduced absorption at zero magnetic field for sample c. For summary of peak energies see Table I.

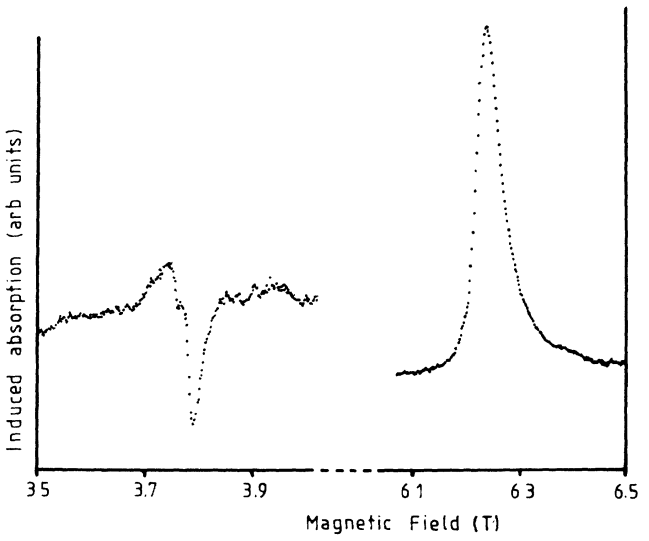

FIG. 2. Photoinduced magnetoabsorption spectra for sample b taken with the 118 μm FIR laser line, showing the inverted $1s$-$2p^+$ donor transition and cyclotron resonance peaks.

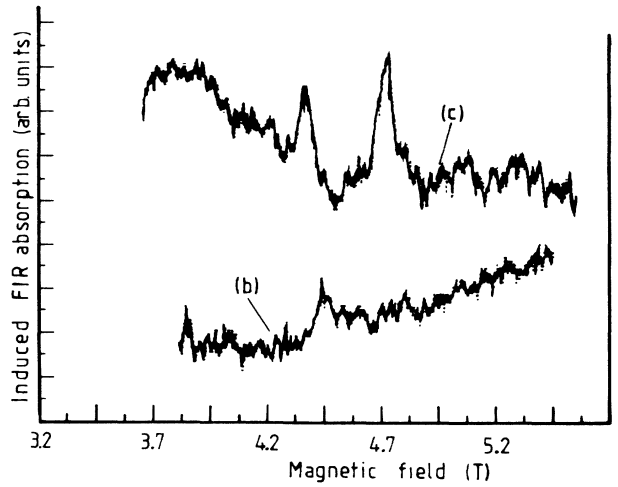

FIG. 3. Type-II excitonic $1s$-$2p$ transitions in the magneto-induced absorption spectra of samples b and c. Observed with the 70.5-μm FIR line.

PTIS donor spectra for high-purity GaAs. For example, the sample of Ref. 11, although showing a $1s$-$2p$ peak width of only ≈ 23 mT (compared with the ≈ 43 mT measured in our samples) and a wealth of higher-order transitions, showed no structure between 3.7 and 5.2 T.

The above considerations, coupled with the fact that the signs of the peaks around 4.5 T in the 70.5-μm magneto-PFA spectrum are the same as the CR peaks, but opposite to the $1s$-$2p$ magnetodonor peaks lead us to conclude that these extra features cannot be due to any GaAs impurity states and we thus assign them to transitions between the states of the type-II excitons in the superlattice layers.

The measured splitting energy between the two peaks in the $B=0$ spectra of samples a and c [although very large compared with the exchange-correlation energy of ≈ 2 μeV measured in optically detected magnetic resonance (ODMR) studies[12] of similar samples] is of the right order of magnitude to be attributed either (i) to inhomogeneous splitting of the $1s$-$2p$ excitonic transition due to interface fluctuations or (ii) to the difference between $1s$-$2p$ and $1s$-$3p$ excitonic transition energies. In the 2D limit the latter two transition energies are expected to differ by a factor of ≈ 1.08 (Ref. 13), in reasonable agreement with the data, but in this case the 2D magnetoexciton theory outlined below predicts magnetic-field shifts ≈ 4 times larger than we observed for the higher-lying line in sample c and we therefore tentatively propose that the splitting originates from interface fluctuations.

The observed splitting energies are in good quantitative agreement with the calculated binding energy shifts arising from fluctuations in the heterojunction positions by one monolayer[14] and further experiments are currently in progress to identify the splitting mechanism with greater certainty. The single broad peak seen in sample b at $B=0$ probably originates from the same effect, with the higher energy line being just unresolved due to the combined effects of inhomogeneous broadening and poor signal-to-noise ratio on the spectra.

The exciton binding energy values (Table I) were calculated on the basis of these assignments, using the results of the $B=0$ 2D excitonic theory of Ref. 13 in which the excitonic Rydberg is given as 1.125 times the $1s$-$2p$ transition energy. The $B=0$ spectral width of each transition in samples a and c is similar to the ≈ 0.5-meV lifetime-limited linewidths (arising from exciton scattering processes) reported for heavy-hole excitons in type-I quantum-well structures[15] and could therefore be entirely homogeneous in origin.

The large magnetic-field shifts of the excitonic transition energies observed provide direct experimental evidence that in these samples the excitons are not being formed from electrons in the X_{xy} minima (i.e., those minima with momenta in the plane of the layers and an in-plane electron mass of $\approx 1.2m_0$). Even in the high-field 2D limit (where the $1s$-$2p^+$ transition becomes an inter-Landau-level transition) and assuming an in-plane hole mass considerably reduced[16-18] to $\approx 0.22m_0$ by valence-band decoupling in the narrow GaAs layers, the transition would move at only 0.63 meV T^{-1} for X_{xy} states. At ≈ 4.5 T though, the ratio of the zero-point cyclotron energy to the X_{xy} excitonic Rydberg would be only ≈ 0.072 resulting in $1s$-$2p^+$ X_{xy} magnetoexciton transition energy shift much less than the 2.1–4.5 meV at ≈ 4.5 T we find. Accordingly we conclude that our excitons have AlAs X_z electronic character, consistent with recent Stark-effect,[19] luminescence decay,[20] luminescence spectroscopy,[21] and optically detected magnetic resonance[12,22] studies.

Recently published finite square-well variational calculations[14] (assuming electrons in the X_z minima, with light masses of $\approx 0.19m_0$ in the layer planes) find binding energies ≈ 10–15% higher than previous infinite barrier models[23,24] and are in excellent agreement with the present work. The small ($\approx 4\%$) residual discrepancy probably originates from uncertainties in the AlAs X-minima band parameters.

To analyze the magnetoabsorption data quantitatively we use the only theory (to our knowledge) suitable for the intermediate range of B fields used here, namely that of Ref. 13. This theory was originally formulated for direct magnetoexcitons in the 2D limit for extremely anisotropic bulk materials (and is hence not ideal for this purpose), but a perturbational extension to the quasi-2D exciton case (applicable for $\mu_\parallel^*/\mu_\perp^* \ll 1$) can be made which introduces a simple energy scaling dimensionality factor $D=1-1.62(\mu_\parallel^*/\mu_\perp^*)^{1/3}$, where $\mu_\parallel^*$ and $\mu_\perp^*$ are the in-plane and perpendicular components of the electron-hole reduced masses, respectively. D here varies between 1 and 0.25 in the 2D and 3D limits, respectively.

In our samples the short-period superlattice heterojunction potentials squeeze the exciton states more tightly against the barriers than would be the case for a single type-II heterojunction. This will act to reduce the effective D value below that which would apply to a single heterojunction and compensates for the effects of the type-II band offsets. The exact effective D value thus depends on the superlattice period and is difficult to calculate analytically.

Because of the large uncertainties in the electron and hole in-plane masses for these thin layers we treat $\mu_\parallel^*$ as a fitting parameter (here assumed equal for all three transi-

tions). We proceed by calculating (for a given assumed value of $\mu_{\parallel}^{*}$) semiempirical D values for each of the three $B=0$ transitions in samples b and c [by comparing the predictions of Eq. (3.15) in Ref. 13 with the corresponding measured $B=0$ peak energies in the PFA spectra]. The resulting modified quasi-2D excitonic Rydberg values are then used to calculate $1s$-$2p^{+}$ transition energies at the respective B values measured in the 70.5-μm magneto-PFA spectra. This process is iterated to arrive at a numerically optimized estimate of $\mu_{\parallel}^{*}$ which gives the best fit to all three resonances in the 70.5-μm magneto-PFA spectra.

The fit yields a figure of $\mu_{\parallel}^{*}=0.13m_0\pm 10\%$ and the values for D and $\mu_{\parallel}^{*}/\mu_{\perp}^{*}$ listed in Table I. The magnitudes of these empirical D parameters indicate excitonic states with a predominantly 2D character (giving us added faith in the value of this approach), and the expected trend of decreasing D with increasing GaAs well width was found. Using the decoupled valence-band value of $\approx 0.22m_0$ (Refs. 16–18) for the in-plane hole mass and $0.19m_0$ (Ref. 25) for the transverse X-point mass in AlAs gives a theoretical $\mu_{\parallel}^{*}$ value of ≈ 0.10, in reasonable agreement with the results of this fitting process.

In conclusion, a spectroscopic measurement of low-dimensional excitonic energy-level structure has been made for the first time. This has allowed us to directly measure the excitonic binding energy term in this important class of structures and we attribute the small ($\approx 4\%$) discrepancy between our measured binding energy values and the variational predictions[14] largely to uncertainties in the AlAs conduction-band parameters used in the latter. The success of this technique opens up a new field of high-resolution low-dimensional exciton-energy-level spectroscopy studies.

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