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## Fano resonances in the excitation spectra of semiconductor quantum wells

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Fano resonances are observed in the photoluminescence excitation spectra of GaAs-AlAs quantum wells. They originate from the Coulomb interaction that couples a discrete state and a continuum of excitonic states belonging to different pairs of conduction and valence subbands. This interpretation is supported by our calculations and by the behavior of the excitonic line shape in the presence of a magnetic field applied perpendicular to the layers.

A Fano resonance refers to the asymmetric line shape in an optical spectrum arising from the interaction of a discrete excited state with a degenerate continuum of states.<sup>1</sup> This asymmetry is produced by the interference between the optical transition amplitudes from a common ground state to a set of coupled excited states of the system. A classical example of such a Fano resonance is provided by the absorption line shapes of autoionization levels that lie between the two ionization thresholds of rare-gas atoms.<sup>2</sup> Typical Fano line shapes were also found in the Raman-scattering spectra of heavily doped *p*-type Si (Ref. 3) and in delta-doped *n*-type GaAs structures<sup>4</sup> as evidenced by the broadening and distortion of the LO-phonon line in the presence of a continuum of electronic excitations.

In semiconductor quantum-well (QW) structures, excitonic features play a prominent role both in the absorption and emission spectra.<sup>5</sup> Excitons are often thought of as quasi-two-dimensional (2D) hydrogen atoms embedded in a high dielectric medium. Their excitonic spectra are analogous to that of a Rydberg series. Indeed, fine structure due to the 2s and 3s excited states of the heavy- and light-hole excitons has been observed in the excitonic spectra of  $Al_xGa_{1-x}As$ -GaAs QW's.<sup>6</sup> With a combination of magnetic and electric fields, excitonic states with high in-plane angular momenta  $(m \ge 1)$  were also reported in one heterostructure of high quality.<sup>7</sup> In a first approximation that assumes parabolic in-plane dispersion and separable wave function for the z and in-plane coordinates, excitons associated with pairs of conduction and valence subbands are uncoupled. Exciton states originating from higher subbands are actually degenerate with the continuum of states of the lower subbands. Thus, a favorable situation may develop for the observation of a Fano resonance in the excitonic spectrum when the coupling between a discrete excitonic state and the continuum of other excitons is sufficiently strong.<sup>8</sup> Although a Fano resonance was predicted when the ground state of the light-hole exciton lies in the heavy-hole continu-um,<sup>9,10</sup> its existence has so far eluded experimental observation.

In this paper we report on the experimental observa-

tion of excitonic line shapes typical of a Fano resonance in the photoluminescence excitation (PLE) spectrum of several quantum-well structures. These strongly asymmetric line shapes offer a striking evidence for the Coulomb coupling between a discrete excitonic state lying within the continuum of the fundamental heavy-hole exciton. Our calculations corroborate the Fano resonance's line shape of these transitions. We also confirm the nature of the Fano resonances by applying a magnetic field perpendicular to the quantum wells. Under these conditions, the continuum of states splits into discrete excited excitonic states, thereby resulting in a progressively vanishing Fano resonance. The simultaneous observation of both an asymmetric line shape and its disappearance in a magnetic field provide a clear identification of a Fano resonance in the optical absorption spectrum of a quasi-2D semiconductor.

We have studied several undoped GaAs-AlAs multiple quantum-well structures (MQW's) grown by molecularbeam epitaxy on a [001]-oriented semi-insulating GaAs substrate. Each structure consists of 90 periods of a double quantum-well unit with nominal thicknesses 48 and 165 Å (or 155 and 160 Å). The width of the AlAs layer separating the two wells remains equal to 14 Å. Each unit is isolated from the previous one by a 100-Å AlAs layer. The sample was mounted on the cold finger of a liquid-helium cryostat and kept at 4.2 K in all the PLE spectra obtained without a magnetic field. The magnetooptical spectra were obtained while the sample was immersed in liquid-helium 3 and was maintained at a temperature of 0.35 K. All spectra were recorded with unpolarized light using either a Ti-sapphire laser or a styryl 9 dye laser pumped by an Ar-ion laser. The laser output power was actively stabilized and kept at a constant level with less than 1% variation over the whole wavelength range.

In Fig. 1 we display the PLE spectrum of the sample that consists of two coupled quantum wells with nominal widths of 165 and 48 Å. The PLE spectra of the other samples were essentially identical and featured a systematic energy shift of the excitonic peaks corresponding to the width variation of the wider of the two wells. The



FIG. 1. Photoluminescence excitation spectrum for a GaAs-AlAs multiple quantum-well sample with 165- and 48-Å-wide wells at 4.2 K. The main excitonic peaks are indicated by arrows and labeled with the principal quantum numbers of the valence and conduction subbands. The inset shows an expanded view of the spectrum below 1.570 eV. Fine excitonic features are described in the text; label 6 indicates the Fano line shape of the exciton state, 1s:hh3-cb1. The spectrometer was set at the maximum of the photoluminescence peak; see arrow labeled (hh1-cb1) at 1.5305 eV.

exceptional quality of the sample is demonstrated by three key elements: the absence of an energy Stokes shift, a full width at half maximum of the ground state of the light-hole exciton of 0.85 meV, and the occurrence of well-resolved excited states of the light- and heavy-hole excitons. In the inset of Fig. 1, peak 1 is the ground state of the light-hole exciton. The peaks labeled 2 and 4 are assigned to the n=2 excited states of the heavy- and light-hole excitons, respectively. They correspond to an exciton state with a circularly symmetric radial envelope wave function since only those states have a finite oscillator strength for a parity-allowed interband transition. The energy separating the 2s excited state from the 1s ground state allows a very precise determination of the exciton binding energy. The experimental value of 7.1 meV (8.7 meV) for the heavy (light) -hole exciton is in excellent agreement with the calculated values for a 165-Å GaAs well surrounded by AlAs confining layers.<sup>11</sup> The theoretical values for E(1s) - E(2s) are 7.3 and 8.2 meV, and correspond to a binding energy of 9.2 and 11.1 meV for the heavy- and light-hole excitons, respectively. As remarked by Andreani and Pasquarello,<sup>11</sup> the larger binding energies in the binary system are a consequence of the large dielectric mismatch between the heterolayers. Owing to the high quality of our sample the 3s excited state of the heavy-hole exciton is also clearly resolved in the PLE spectrum as a separate line at 1.5396 eV (peak 3 in inset). Its identification will be discussed in relation to the magneto-optical data of Fig. 2. Line 5 is assigned to the parity-forbidden 2p exciton transition between n=2heavy-hole subband and n = 1 electron subband. Its finite oscillator strength originates from a Coulomb coupling



FIG. 2. Calculated Fano-like line shape of the excitonic absorption associated to the first electron subband and third heavy-hole subband in a double quantum-well structure. The continuum absorption is normalized to one away from the coupling region. The peak of the resonance is predicted to occur at 1.5587 eV in excellent agreement with experimental position, 1.5586 eV.

between the 1s:lh1-cb1 excitonic state and a 2p:hh2-cb1 excited state that transfers to the latter a finite amount of oscillator strength.<sup>8,12</sup> We note that this transfer can only occur if valence-band mixing is considered. The ground state of this parity-forbidden exciton is not normally observed unless the inversion symmetry of the confining potential is broken by the application of an external electric field along the growth direction.<sup>13</sup>

In the remaining part of this paper, we will concentrate our attention on a remarkable feature of our PLE spectrum that spans an energy range between 1.550 and 1.560 eV (line labeled 6 in inset of Fig. 1). The maximum intensity occurs at an energy of 1.5586 eV. Its line shape is highly asymmetric unlike other excitonic lines in this excitation spectrum. Furthermore, the absorption is strongly suppressed on the low-energy side of the line while on the high-energy side it recovers to a level indicated by a dotted line in the inset of Fig. 1. This straight dotted line corresponds to the absorption level of a 2D continuum of energy states in the absence of any excitonic effects. A highly asymmetric line shape is suggestive of a Fano resonance that may happen when a discrete exciton state is degenerate with the continuum of states of other excitonic transitions. We assign this excitonic feature to the states derived from the n = 1 electron subband and the n = 3 heavy-hole subband based on our calculation of the exciton transitions for a 165-Å well and an exciton binding energy of 7.5 meV. We emphasize that this excitonic state lies in the continuum from both the light- and heavy-hole excitons that originate from the lowest conduction and hole subbands.

It is important to consider an alternative effect that may decrease the absorption in the vicinity of the hh3cbl exciton: this effect arises from the correlation between an unbound electron-hole pair lying in the continuum of states of the heavy (alternatively light) -hole exciton. In consequence, the absorption coefficient has to be corrected by an enhancement factor known as the Sommerfeld factor. In a 2D system, it is given by<sup>14</sup>

$$S(E) = 2/(1 + e^{-\pi\sqrt{E_b/E}})$$
,

where  $E_b$  is the exciton binding energy and E the kinetic energy of the electron-hole pair. This factor varies between 2 and 1 on an energy scale given by the binding energy; moreover, a realistic estimation of the enhancement factor for a quasi-2D system yields a value between 1.3 and 1.4 at the subband edge.<sup>15</sup> A simple estimation of S(E) between  $E = E_b$  and  $E = 2E_b$  yields a relative variation of the absorption coefficient of 6%, which is quite insufficient to account for the experimental value of about 20%. Thus, the observed absorption minimum on the low-energy side of the hh3-cb1 line is a definitive signature for a Fano resonance.

The exciton, 1s:hh3-cb1, even though derived from a valence and conduction subband with different quantum numbers n, has a nonzero oscillator strength because the overlap integral of the envelope wave functions in the z direction takes a finite value for a finite potential barrier. On the one hand, the binary compound in the barrier favors narrow linewidths in the excitation spectrum by preventing the broadening from the alloy scattering. On the other hand, the double quantum-well structure allows a higher penetration of the heavy-hole wave function into the barrier resulting in a sizable oscillator strength for the hh3-cb1 transition despite the high potential barriers.

As already mentioned above, a Fano resonance originates from the coupling between a discrete excitonic state and an excitonic continuum associated to a pair of lower subbands. We have identified two candidates for this coupling mechanism: The first one corresponds to the off-diagonal part of the Luttinger-Kohn Hamiltonian, which is responsible for valence-band coupling; the second one originates from the 3D character of the Coulomb interaction. While the former interaction couples the 1s:hh3-cb1 exciton with the light-hole exciton states with p,d radial symmetries that are derived from different pair of subbands, it cannot give rise to the observed interference because exciton states with radial symmetries other than s are not optically active. Coupling to these states will only contribute to an energy shift of the excitonic line and its broadening owing to its sharing of oscillator strength with the continuum states close in energy to the peak of absorption. Let us emphasize that only the second mechanism can possibly give rise to a suppression of absorption on the low-energy side of the 1s:hh3-cb1 excitonic line. This interaction couples the heavy-hole excitonic states derived from separate valence subbands provided that the excitonic states have the same radial symmetry; thus, 1s:hh3-cb1 exciton is coupled to all the continuum excitonic states derived from hh1 and cb1 subbands whose envelope wave functions have s-radial symmetry. In Fig. 2, we present the results of a calculation of the Fano resonance's line shape for that excitonic transition.<sup>16</sup> As can be seen, the existence of a negative interference on the low-energy side of the excitonic line is confirmed by these results. The position of the resonance is also accurately determined by this calculation. On the other hand, the calculated resonance line shape only provides a qualitative comparison

with the experimental data. This discrepancy probably arises from the contribution of interface roughness and impurities to the exciton linewidth.

The density of states of a two-dimensional electron system is known to become fully discrete by the application of a magnetic field. This profound modification of the energy spectrum was utilized to investigate the behavior of the Fano resonance. In Fig. 3, we display a set of PLE spectra in the presence of a magnetic field applied perpendicularly to the layers. Modification of the PLE spectrum became noticeable at a magnetic field exceeding 0.5 T. The oscillator strength of the 1s-lh exciton is gradually reduced in favor of excited excitonic states with higher quantum numbers. In particular, the 2s and 3s magnetoexcitons with mostly heavy-hole character acquire an oscillator strength nearly as large as that of the ground state of the light-hole exciton. Assignment of these states is corroborated by their behavior in a magnetic field: Thus, the magnitude of the energy shift for the 2s exciton is well explained by the diamagnetic shift of a 2D hydrogenlike exciton,<sup>17</sup> while that of the 3s exciton was not as large as predicted presumably because of the effect of valence-band mixing. Above 0.5 T new magnetoexciton lines appear on the low-energy side of the Fano resonance. Note that up to six new lines are superimposed in this part of the spectrum at a field of 0.9 T. Furthermore, the application of a magnetic field does not lead to the immediate disappearance of the Fano resonance. Up to a field of 1 T, the continuum background absorption is modulated by the emergence of the new magnetoexcitons. At a field of 1.1 T, the Fano resonance is still clearly observed; the remaining interference with the continuum of



FIG. 3. Influence of a magnetic field applied perpendicular to the layers on the PLE spectrum at 0.35 K. The Fano line shape of the 1s:hh3-cb1 exciton is completely suppressed by a field of 1.2 T. New magnetoexcitons emerge from the absorption background as the field reaches 0.9 T. Peaks that are labeled on upper spectrum correspond to the first four excitonic states with *s*-radial symmetry in inset of Fig. 1.

states takes the form of a trough at 1.555 eV. Beyond 1.2 T, the Fano resonance completely disappears and is replaced by a plethora of new lines reflecting the discreteness of the energy spectrum.

The observed modification of the PLE spectrum upon application of a magnetic field provides important evidence supporting the identification of the Fano resonance. An identification of the new magnetoexciton lines is easily achieved based on symmetry arguments.<sup>10</sup> The new lines arise from excited magnetoexcitonic states that acquire a significant oscillator strength due to mixing with nearby optically active excitons. In the presence of a magnetic field, strong mixing occurs between the 2sexcited state from the light-hole magnetoexciton and heavy-hole exciton states with d-radial symmetry and principal quantum number 3 when the field moves the lines closer in energy as evidenced in a fan diagram.<sup>18</sup> Thus, a pair of lines originate from this coupling as the magnetic field increases. Similarly, a pair of lines from the light-hole magnetoexciton state with d-radial symmetry can also draw a significant oscillator strength from its coupling with the 3s-excited state of the heavy-hole exciton. The other two lines may be the magnetoexciton states labeled 5 (i.e., 2p:hh2-cb1) in the inset of Fig. 1 or a pair of magnetoexciton states with principal quantum number 4 and *d*-radial symmetry. We note that magnetoexcitons evolving from states with higher principal quantum numbers are expected to have a weaker oscillator strength. Other excited *s*-like excitonic states were also considered but are not expected to contribute because a calculation of their energy shift in a magnetic field assuming pure hydrogeniclike excitonic states yields a much larger dependence than experimentally observed.

In summary, we have reported the observation of a typical Fano resonance in the PLE spectrum of MQW's. The resonance was attributed to the interference between the discrete excitonic state 1s:hh3-cb1 and the continuum of excitonic states evolving from the first heavy-hole and conduction subbands. This was confirmed by a calculation of the Fano resonance's line shape of this excitonic transition. Additional evidence was obtained from the suppression of the Fano line shape by the application of a magnetic field. The occurrence of a Fano resonance demonstrates the importance of the Coulomb interaction in coupling exciton states derived from different pairs of subbands.

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- <sup>1</sup>U. Fano, Phys. Rev. **124**, 1866 (1961).
- <sup>2</sup>H. Beutler, Z. Phys. **93**, 177 (1935).
- <sup>3</sup>F. Cerdeira, T. A. Fjeldly, and M. Cardona, Solid State Commun. **13**, 325 (1973); Phys. Rev. B **8**, 4734 (1973).
- <sup>4</sup>L. A. O. Nunes, L. Ioriatti, L. T. Florez, and J. P. Harbison, Phys. Rev. B **47**, 13 011 (1993).
- <sup>5</sup>R. Dingle, W. Wiegmann, and C. H. Henry, Phys. Rev. Lett. 33, 827 (1974); R. C. Miller, D. A. Kleinman, W. T. Tsang, and A. C. Gossard, Phys. Rev. B 24, 1134 (1981); R. C. Miller and D. A. Kleinman, J. Lumin. 30, 520 (1985), and references therein.
- <sup>6</sup>D. C. Reynolds, K. K. Bajaj, C. Leak, G. Peters, W. Theis, P. W. Yu, K. Alavi, C. Colvard, and I. Shidlovsky, Phys. Rev. B 37, 3117 (1988).
- <sup>7</sup>L. Viña, G. E. W. Bauer, M. Potemski, J. C. Maan, E. E. Mendez, and W. I. Wang, Phys. Rev. B 38, 10154 (1988).

- <sup>8</sup>H. Chu and Y. C. Chang, Phys. Rev. B 39, 10861 (1989).
- <sup>9</sup>D. A. Broido and L. J. Sham, Phys. Rev. B 34, 3917 (1986).
- <sup>10</sup>G. E. W. Bauer and T. Ando, Phys. Rev. B 38, 6015 (1988).
- <sup>11</sup>L. C. Andreani and A. Pasquarello, Europhys. Lett. 6, 259 (1988); Phys. Rev. B 42, 8928 (1990).
- <sup>12</sup>L. W. Molenkamp, G. E. W. Bauer, R. Eppenga, and C. T. Foxon, Phys. Rev. B 38, 6147 (1988).
- <sup>13</sup>D. A. B. Miller, J. S. Weiner, and D. S. Chemla, IEEE J. Quantum Electron. 22, 1816 (1986).
- <sup>14</sup>M. Shinada and S. Sugano, J. Phys. Soc. Jpn. 21, 1936 (1966).
- <sup>15</sup>K. S. Chan, J. Phys. C 20, 791 (1987).
- <sup>16</sup>For a complete description of the calculation, see J. A. Brum and D. Y. Oberli, in Proceedings of the International Conference on Excitons, Montpellier, 1993 [J. Phys. Paris Colloq. C5-3, 191 (1993)].
- <sup>17</sup>A. H. MacDonald and D. S. Ritchie, Phys. Rev. B **33**, 8336 (1986).
- <sup>18</sup>D. Y. Oberli (unpublished).