Quantum Confined Fano Interference

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We study the transition from a dense continuum to a sparse quasicontinuum in the Fano problem. Transmission measurements on epitaxial layers of GaAs in a high magnetic field and calculations of the optical absorption show how the Fano interference disappears as quantum confinement discretizes the continuum states. The transition between quasi-one-dimensional and quasi-zero-dimensional systems occurs at length scales which are unusually large for optical experiments. [S0031-9007(97)02443-5]

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Quantum size effects have been thoroughly investigated since advances in material synthesis made possible the growth of semiconductor heterostructures with atomic monolayer accuracy. They become apparent in a physical phenomenon when at least one dimension of the sample becomes comparable to or smaller than the length scale that governs the quantum mechanics of that phenomenon. In the optical domain the exciton Bohr radius a_o is usually the relevant length scale, and quantum confinement dominates the optical properties of semiconductor structures of size $L \leq a_o$ [1,2]. In the case of transport the carrier de Broglie wavelength λ_B determines the length scale at which quantum transport effects are observed [3]. In this Letter we present a new example of quantum size effects, apparent in the line shape of an optical resonance involving the quantum mechanical coupling between a localized, discrete state and a quasicontinuum. As shown below, the size effect is a direct consequence of the energy quantization due to the finite sample thickness. Remarkably, it appears for length scales $L > 15a_o$ that are unusually large for the optical domain.

Quantum mechanical interference between a discrete state and a quasicontinuum is a fundamental problem in physics. It is one of a few exactly solvable models in many-body theory [4], and as such it gives insight into other many-particle phenomena. This problem was simultaneously formulated by Anderson and Fano. Anderson applied it to the mixing of localized impurity orbitals and extended conduction band states in solid state physics [5]. Fano used it to describe the interference of electronic transitions in atomic spectra [6]. The coupling of the discrete state to the quasicontinuum in the *Fano-Anderson model* is, furthermore, a prototype of irreversible decay [7,8]. It can represent various fundamental relaxation mechanisms, such as spontaneous emission, nuclear decay, the autoionization of excited atoms originally discussed by Fano [6], intramolecular relaxation, and coupling to a phonon bath [9]. The quantum interference manifests itself as a resonance in the optical absorption spectrum with a characteristic asymmetric line shape. Very close to its peak, a pronounced minimum occurs at an energy where the transition amplitudes of the discrete state and the continuum interfere destructively. It has been observed in a variety of atomic, molecular, and semiconductor systems [10], and recently in the optical absorption of bulk semiconductors in high magnetic fields [11]. In the latter case, the coupling between degenerate one-dimensional continuum states and magnetoexcitons of different Landau indices was shown to result from the Coulomb interaction [11].

An important underlying assumption of the Fano solution is that the manifold of states is sufficiently dense to be treated as a continuum. In many physical situations, however, the manifold of closely spaced energy levels is rather sparse. Such is the case in molecular spectroscopy, where radiationless relaxation of electronic transitions in polyatomic molecules occurs due to the interaction with the molecule's vibrational spectra. This case was analyzed by Bixon and Jortner [12] using a formalism analogous to the one used by Fano in his treatment of atomic autoionization [6]. Although the two cases of dense and sparse quasicontinua have been analyzed, the smooth transition between them has been ignored due to the lack of a relevant physical situation. However, modern semiconductor growth techniques allow tuning of the level spacing in thin epitaxial layers by means of tailoring the layers' thickness and potential discontinuities. When applied to the one-dimensional quasicontinua of magnetoexcitonic Fano resonances [11], the tuning of level spacing results in a transition from a dense to a sparse manifold as a function of sample thickness.

In this paper we study the transition from the dense continuum of the Fano-Anderson model to a sparse quasicontinuum of closely spaced discrete energy levels, as the sample thickness is decreased. Transmission measurements on thin epitaxial layers of GaAs in high magnetic field show that the Fano interference disappears as quantum confinement discretizes the quasicontinuum of states. To support our experimental findings we calculate the optical absorption in the framework of the "picket fence" model [12,13], i.e., a quasicontinuum of equidistant levels with identical oscillator strengths. We derive an analytic expression for the absorption spectrum which lends itself

to a straightforward numerical study of the evolution of the line shape. The calculations are in good agreement with the experimental data. Thus our work demonstrates a unique transition between quasi-one-dimensional and quasi-zero-dimensional systems. Remarkably, this transition occurs already for weak confinement, i.e., for length scales of the confining potential which are much longer than the exciton Bohr radius. This establishes a new length scale in semiconductor optics.

To demonstrate the effect of quantum confinement on the Fano resonance, we present experimental absorption spectra in high magnetic field, measured on a set of high quality GaAs samples. Each of the three samples, all grown by molecular beam epitaxy, has a GaAs layer of different thickness sandwiched between AlGaAs layers. The GaAs layer thicknesses are $L = 1$ μm, $L = 0.5$ μm, and $L = 0.25$ μm. All three samples are antireflection coated on both sides, and glued on c-axis sapphire. The GaAs substrates were removed by selective chemical etching to allow transmission measurements. The samples are placed at $T = 1.6$ K in a $B \leq 12$ T magneto-optical cryostat. The split-coil magnet allows measurements with the field applied either perpendicular to the sample surface or in plane with it. Light from an incandescent lightbulb is passed through a polarizer and a quarter-wave plate before being focused on the sample, to allow measurements with circular polarizations. The transmitted light is dispersed in a spectrometer and collected in an optical multichannel analyzer. The absorption spectra are calculated from the normalized transmission measurements.

Figure 1 presents the absorption spectra of the three samples in a magnetic field $B = 6$ T applied perpendicular to the sample surface. These measurements were performed using σ^- light polarization. Each trace shows two series of magnetoexcitonic resonances, associated with heavy-hole and light-hole transitions. The lowest heavy-hole and light-hole magnetoexcitons have Lorentzian shapes, and their degeneracy is removed by the combined effects of mechanical strain [14] and different diamagnetic shifts. The higher order magnetoexcitons in all three samples coincide and have clear asymmetric profiles. However, only the data from the 1 and 0.5 μm samples show clear Fano line shapes which dip below the continuum absorption on their high-energy sides. The dips are most pronounced in the spectrum measured with the 1 μm sample, whereas they are completely absent from that of the 0.25 μm sample.

The most plausible explanation for the behavior seen in Fig. 1 is the suppression of the Fano interference due to a quantum size effect. As the sample thickness is decreased, confinement by the potential discontinuities forms a set of closely spaced subbands. In the presence of a magnetic field this results in discretization of the one-dimensional continuum which is responsible for the Fano interference [11]. The fact that this occurs for relatively thick samples is intriguing, since quantum size effects in optical experiments are usually observed at length scales

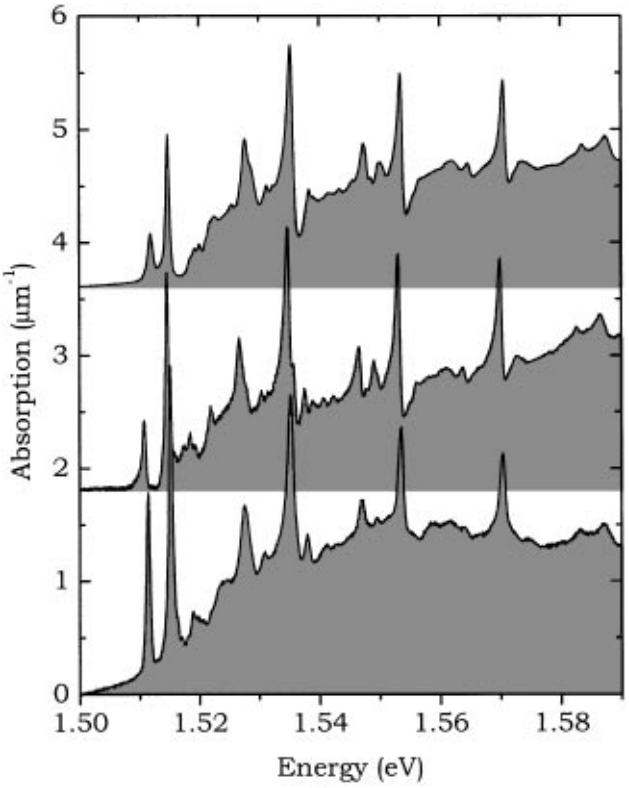

FIG. 1. Absorption spectra of three thin GaAs samples in a magnetic field of 6 T applied perpendicular to the sample surface, for σ^- light polarization. Top to bottom: 1 μm sample; 0.5 μm sample; 0.25 μm sample.

of the order of a *few* nm. However, a simple calculation shows that the energy level spacing in the quasicontinuum becomes comparable to the typical Fano coupling strength Γ (manifested as the width of the Fano resonance [6]) for quantum wells as wide as 1 μm. In this case the line shape would no longer be dominated by the coupling to the continuum. To support this interpretation we have repeated our measurements with the magnetic field applied in the plane of the sample. In this case the free motion of the carriers along the direction of the applied field is not quantized by the potential discontinuities of the sample. Moreover, the confinement by the potential discontinuities can be neglected as long as the magnetic length l_H is small compared to the sample thickness. In a field of 10 T $l_H \approx 8.1$ nm, and this condition is satisfied for all three samples. We would thus expect all samples to show identical absorption spectra under these conditions. However, the absorption spectra do not retain the simple form of Fig. 1 in this configuration, since the optical selection rules become rather complicated even for circularly polarized light [15]. Figure 2 shows the absorption spectra of the 0.25 and 0.5 μm samples in a parallel field of 10 T. The spectra are far more complicated than those in Fig. 1, and none of the samples show any clear Fano resonances. We point out, however, that the two traces in Fig. 2 are virtually identical, as expected. This rules out the possibility that the clear

differences shown in Fig. 1 are due to any extrinsic effect, and confirms our interpretation (incidentally, the narrower lines seen in the data from the 0.25 μm sample indicate that its quality is higher).

As mentioned before, a simple calculation of the energy level spacing in a quantum well shows that discretization of the continuum of states above each magnetoexciton becomes relevant for sample thicknesses below 1 μm. In order to study how this discretization affects the Fano line shape, we calculate the optical absorption in the framework of the "picket fence" model [12,13] shown in Fig. 3. $|x\rangle$ is a discrete excitonic energy level, and the states $|k\rangle$ form a quasicontinuum. The energies of the quasicontinuum states are $\epsilon_k = k\Delta\epsilon$, where k is an integer. The ground state is denoted by $|g\rangle$. The Hamiltonian of the system is

$$H = \epsilon_x|x\rangle\langle x| + \sum_k \epsilon_k|k\rangle\langle k| + \\ + \sum_k (V_k|k\rangle\langle x| + V_k^*|x\rangle\langle k|)\,, \qquad (1)$$

where V_k is the coupling between the states $|x\rangle$ and $|k\rangle$. This Hamiltonian is identical to the Fano-Anderson model [5,6], with the exception that the states $|x\rangle$ and $|k\rangle$ describe one-photon excitations of the semiconductor rather than single electron states. In the Fano model it is assumed that V_k is slowly varying in the vicinity of the discrete level ϵ_x, and that the quasicontinuum is dense, allowing replacement of the summation with explicit integration and a straightforward solution [6]. We, however, keep the discrete summation and use the Green's function formalism. The Dyson equation $G = G^{(0)} + G^{(0)}VG$ for the excited states gives a system of coupled equations for G_{xx} and G_{kx} [13]. As in the Fano model, we assume that the only nonzero matrix elements of the coupling are $V_{kx} = V_{xk} = V$. Solving for G_{xx} we get

$$G_{xx} = \left[G_{xx}^{(0)-1} - V^2 \sum_k G_{kk}^{(0)} \right]^{-1}. \qquad (2)$$

$G_{xx}^{(0)-1} = \epsilon - \epsilon_x + i\eta$, where η is a small positive number, interpreted as the natural linewidth. We assume, for simplicity, the same natural linewidth η for the discrete and picket fence states, so that $G_{kx} = G_{xk} = VG_{xx}/(\epsilon - k\Delta\epsilon + i\eta)$, and

$$\sum_k G_{kk}^{(0)} = \frac{1}{\Delta\epsilon} \sum_k \frac{1}{\frac{\epsilon+i\eta}{\Delta\epsilon} - k} \\ = \frac{\pi}{\Delta\epsilon} \cot\left[\frac{\pi(\epsilon + i\eta)}{\Delta\epsilon} \right]. \qquad (3)$$

From the Dyson equation we also find $G_{kl} = G_{kk}^{(0)}\delta_{kl} + V^2 G_{xx} G_{kk}^{(0)} G_{ll}^{(0)}$. The optical absorption is given by the optical theorem [16]: $\sigma_g(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\{T_{gg}(\epsilon_g + \omega)\}$ where $T = \mu + \mu G \mu$ is the T matrix, μ is the dipole moment operator, and the index g refers to the ground state $|g\rangle$. We express T_{gg} in terms of the matrix elements of the Green's function

$$T_{gg} = \mu_{gx} G_{xx} \mu_{xg} + \sum_k [\mu_{gk} G_{kx} \mu_{xg} + \mu_{gx} G_{xk} \mu_{kg}] \\ + \sum_{kl} \mu_{gk} G_{kl} \mu_{lg}\,. \qquad (4)$$

For simplicity, we assume, as usual, that μ_{kg} is independent of k, and express T_{gg} in terms of $a \equiv \mu_{xg}/\mu_{kg}$. We can further assume, without loss of generality, that a is real. We get the simple expression

$$T_{gg} = |\mu_{gx}|^2 \left[G_{xx} \left(1 + \frac{V}{a} \sum_k G_{kk}^{(0)} \right)^2 \right. \\ \left. + \frac{1}{a^2} \sum_k G_{kk}^{(0)} \right], \qquad (5)$$

where the last term is the background contribution due to the manifold of states $|k\rangle$. We finally arrive at an analytic expression for the absorption

$$\sigma(\omega) = -\frac{|\mu_{gx}|^2}{\pi}\,\mathrm{Im}\left\{ \frac{[1 + \frac{1}{q}\cot\pi(\frac{\hbar\omega+i\eta}{\Delta\epsilon})]^2}{\hbar\omega - \epsilon_x + i\eta - \Gamma\cot\pi(\frac{\hbar\omega+i\eta}{\Delta\epsilon})} \right. \\ \left. + \frac{1}{q^2\Gamma}\cot\pi\left(\frac{\hbar\omega + i\eta}{\Delta\epsilon} \right) \right\}, \qquad (6)$$

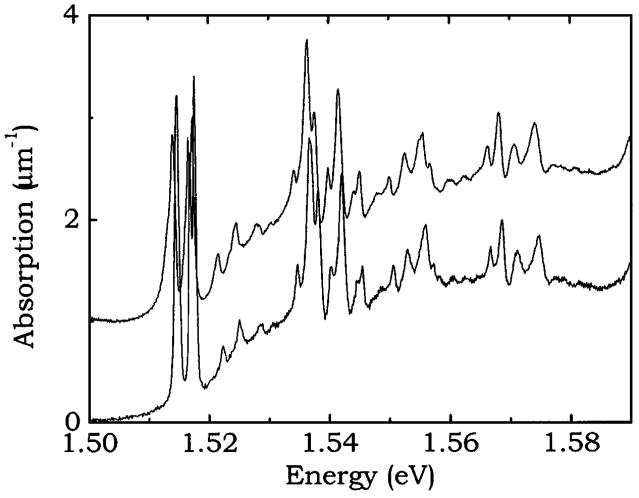

FIG. 2. Absorption spectra of two of the samples in a magnetic field of 10 T applied parallel to the sample surface, for σ^- light polarization: 0.5 μm sample (top), and 0.25 μm sample (bottom).

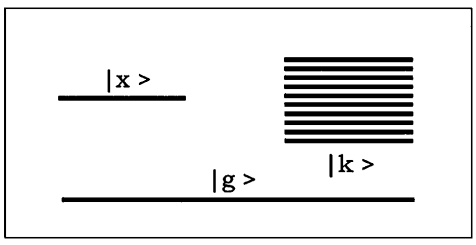

FIG. 3. Energy level scheme for the model described in the text. $|g\rangle$ is the ground state of the semiconductor. $|x\rangle$ is a discrete excited state, coupled to a manifold of quasicontinuum states $|k\rangle$.

where $q \equiv a\Delta\epsilon/\pi V$ and $\Gamma \equiv \pi V^2/\Delta\epsilon$ differ from the equivalent Fano parameters [6] only in the appearance of $\Delta\epsilon$ due to different normalization of the picket fence and continuum states. If we take the "continuum limit," letting $\Delta\epsilon \to 0$ while keeping q and Γ fixed, we recover the exact Fano form in Eq. (6). In this limit, V scales as $\sqrt{\Delta\epsilon}$, a scales as $1/\sqrt{\Delta\epsilon}$, and $\cot \pi(\frac{\hbar\omega + i\eta}{\Delta\epsilon}) \to -i$.

Figure 4 illustrates the results of Eq. (6) for three sets of parameters which correspond to the three samples which we have investigated. In Fig. 4(a) we have used q and Γ parameters obtained from a fit of the Fano line shape [6] to the absorption spectrum of the 1 μm sample shown in Fig. 1. The level spacing used is $\Delta\epsilon \approx 0.8$ meV, corresponding to the spacing between the adjacent one-dimensional "particle in a box" energy levels, with Landau index $m = 0$, around the magnetoexciton with Landau index $n = 1$ at 1.5352 eV. The calculation for this case results in a clear Fano line shape. In Figs. 4(b) and 4(c) the level spacing $\Delta\epsilon$ is increased by factors of 2 and 4 relative to Fig. 4(a), corresponding to the 0.5 and 0.25 μm samples, respectively. As the level spacing is increased the ratio of dipole moments is scaled as $a \propto 1/\sqrt{\Delta\epsilon}$, so that the average oscillator strength of the quasicontinuum is conserved. V is kept constant throughout Fig. 4, in line with the basic Fano model. With these assumptions the Fano parameters scale as $q \propto \sqrt{\Delta\epsilon}$ and $\Gamma \propto 1/\Delta\epsilon$. Figure 4(b) still shows a Fano line shape, attenuated as compared to Fig. 4(a). In contrast, Fig. 4(c) does not exhibit a clear dip on the high energy side of the resonance. The comblike background contribution of the quasicontinuum in Fig. 4(c) results because $\eta < \Delta\epsilon$. The behavior seen in Fig. 4 is in good agreement with the experiment.

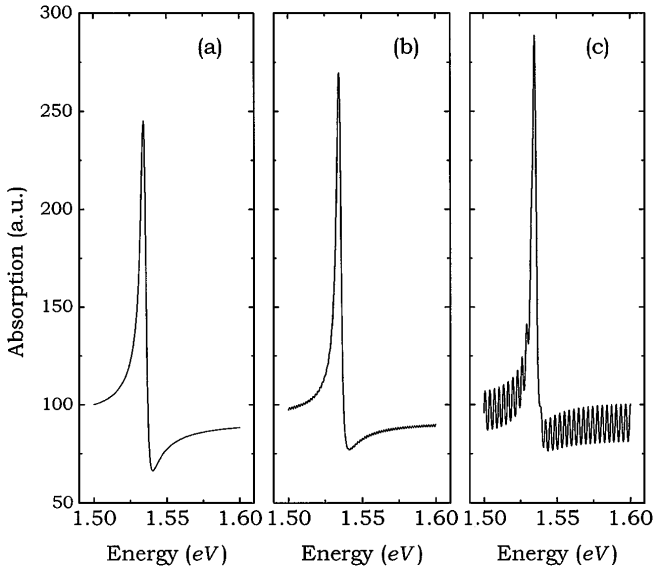

FIG. 4. Numerical evaluations of Eq. (6) for several sets of parameters: (a) $\epsilon_x = 1.5352$ eV, $\Gamma = 0.58$ meV, $q = -2.44$, $\Delta\epsilon = 0.8$ meV, $\eta = 1.5$ meV; (b) $\epsilon_x = 1.5352$ eV, $\Gamma = 0.29$ meV, $q = -3.45$, $\Delta\epsilon = 1.6$ meV, $\eta = 1.5$ meV; (c) $\epsilon_x = 1.5352$ eV, $\Gamma = 0.145$ meV, $q = -4.88$, $\Delta\epsilon = 3.2$ meV, $\eta = 1.5$ meV.

In conclusion, we have shown how the Fano interference disappears as the manifold of continuum states becomes sparse. The experimental demonstration is a new and unique example of a confinement effect, in this case resulting in a transition between quasi-one-dimensional and quasi-zero-dimensional systems, and appearing for length scales unusually large for optical experiments.

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