Fano Interference in Type-II Semiconductor Quantum-Well Structures

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We predict, on the basis of a model calculation, that Fano interference effects may be observed in type-II quantum-well structures resulting in pronounced nonreciprocal absorption and emission profiles and interference narrowing due to overlapping resonances.

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The effect of Fano interferences on the absorption and emission profiles has gained considerable interest recently in respect to what is loosely referred to as "lasing without inversion" [1]. Fano interference [2] occurs when optical transitions couple a quasidiscrete state $|a\rangle$ and an energetically degenerate continuum $\{|c_E\rangle\}$ to a common ground state $|i\rangle$ and when the state $|a\rangle$ is quantum mechanically coupled to the continuum with matrix element V. In such a system the absorption spectrum exhibits a dip with zero absorption while the emission spectrum does not show a corresponding dip if the state $|a\rangle$ is populated (pumped) incoherently. Consequently, there is a frequency range where one has emission, but no absorption [3]. Since these frequencies are situated at the wing of the emission line, the effect is not very pronounced. Harris [1] has shown that an additional quasidiscrete state may produce a dramatic amplification of this effect. Although he has in mind atomic systems he also suggests that in artificially layered materials nonreciprocal line shapes should exist as well.

In this Letter we predict, on the basis of a model calculation, that Fano interferences may be observed in type-II semiconductor quantum-well structures resulting in pronounced nonreciprocal absorption and emission profiles. In particular, we show that interference narrowing of both the emission and absorption lines occurs and that the susceptibility for stimulated emission is much larger than that for absorption.

Even though it seems feasible to observe Fano interference effects in indirect-gap semiconductors where coupling of the direct and indirect electronic states occurs [4–7], the situation is much more transparent in artificially layered semiconductor heterostructures. In particular, type-II GaAs/AlAs quantum-well structures seem to fulfill all the fundamental requirements for the occurrence of Fano interferences. We thus suggest to consider these structures for the so-far missing experimental verification of the principle underlying the proposal by Harris [1].

In the GaAs/AlAs system the well is formed by the direct-gap semiconductor GaAs, whereas the barriers are formed by the indirect-gap semiconductor AlAs. In GaAs both the top of the upper valence band and the bottom of the lowest conduction band are at the center of the Brillouin zone (Γ point). The valence-band structure is similar in AlAs; however, the lowest conduction-band minimum is at the X point and the Γ minimum is at higher energies. The band alignment of a GaAs/AlAs quantum well is depicted in the upper part of Fig. 1 showing the spatial variation of the Γ- and X-band extrema. It should be noted that the $E(\mathbf{k})$ dispersion parallel to the layers leads to energy-band continua above the discrete levels shown. The following characteristic features of the GaAs/AlAs structure are of particular importance [8] for our purposes.

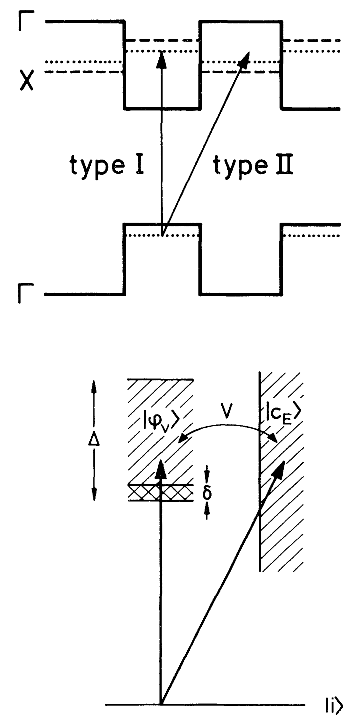

FIG. 1. Band alignment of a type-II GaAs/AlAs quantum-well structure showing the spatial variation of the bulk Γ- and X-band extrema together with the heterostructure band extrema (dotted lines), and schematic representation of the model system (bottom).

(i) For sufficiently thin GaAs layers ($L_z \leq 3.5$ nm) the lowest confined Γ conduction-band state is at higher energy than the lowest X conduction-band state of the AlAs layer. The Γ states then are degenerate with the X continuum of the AlAs. Electrons generated optically in the Γ conduction-band states of the GaAs layer therefore relax into the lower X states of the AlAs layer [9], resulting in a confinement of electrons and holes in different slabs of the structure. This is referred to as a type-II structure in contrast to the type-I structures, where electrons and holes are confined to the same slab.

(ii) Optical transitions without phonon participation are possible into both the Γ (type-I transitions) and X conduction-band states (type-II transitions). The type-II transitions become allowed in the ideal quantum-well structure due to the new symmetry (with respect to the bulk) in the direction perpendicular to the slabs. The type-I transitions, however, dominate the absorption [8,9].

(iii) In real structures the interface roughness leads to coupling between states with probability amplitude predominantly in the GaAs or the AlAs layer, respectively. This interface roughness can couple states with different momentum parallel to the interface. The amount of momentum that can be transferred by this interface roughness depends on its characteristic length scale. This additional coupling manifests itself in a phononless Γ-X scattering taking place on a subpicosecond time scale [9]. For the following discussion it is essential that the mechanism responsible for the optical coupling between the ground state and the "indirect" electron-hole continuum is different from that coupling the Γ- and X-related conduction-band states.

(iv) The optical transitions are strongly affected by excitonic effects. In particular, the type-I transitions close to the effective band gap are enhanced as compared to transition into higher continuum states.

Based on these experimentally verified properties we simulate the type-II GaAs/AlAs quantum-well structure by the model depicted in the lower part of Fig. 1. Optical transitions are possible from the ground state $|i\rangle$ of the semiconductor both to an electron-hole continuum $\{|c_E\rangle\}$ (on the right-hand side of Fig. 1) and to the states $|\varphi_\nu\rangle$ which represent the left-hand continuum. These states, when coupled by some static perturbation to the continuum $\{|c_E\rangle\}$, form a continuous set of overlapping resonances. Mies [10] has studied a similar situation in the context of electron-molecule scattering and found dramatic interference narrowing of line shapes under certain conditions. Experimentally, interference narrowing has been observed, e.g., at crossings of sodium Stark resonances [11].

The strength of the optical transitions into the left-hand continuum relative to that for transitions into the right-hand continuum is denoted by the Fano parameter [2] q_ν. Modeling the type-II structures, the states $|\varphi_\nu\rangle$ correspond to a direct excitonic band. Furthermore, because of the **k** selection rule q_ν is large only for transitions into the lowest states in the interval δ. This interval has a finite width, since the static disorder leads to an uncertainty in the total momentum **K** and thus in energy. The $|\varphi_\nu\rangle$ states are coupled due to interface roughness to the $\{|c_E\rangle\}$ continuum. We assume that this coupling is effective for states in the limited energetic range Δ only and is independent of energy. The excited-state part of the Hamiltonian can then be diagonalized exactly [2], leading to the Fano states $|E\rangle$, which are stationary in contrast to the states $|\varphi_\nu\rangle$ and $|c_E\rangle$.

Calculating the emission and absorption line shapes we follow Agarwal, Haan, and Cooper [12] and write the corresponding susceptibilities in terms of the transient susceptibility $\chi(z,\omega)$,

$$\chi(\omega) = -\lim_{z\to 0} z\chi(z,\omega) ,$$

where for absorption,

$$\chi_{\rm abs}(z,\omega) = \int \frac{v^*(E)}{E_0^2} \rho_{Ei}^{i}(z)\, dE ,$$

and for emission,

$$\chi_{\rm em}(z,\omega) = -\frac{\Gamma}{z} \int \frac{v^*(E)}{E_0^2} \rho_{Ei}^{a}(z)\, dE .$$

The density matrix $\rho_{Ei} = \langle E|\rho|i\rangle$ between the ground state $|i\rangle$ at zero energy and the exact Fano eigenstates $|E\rangle$ is calculated from $\partial_t \rho = -i[H,\rho]$ to first order in $v(E)$ and ρ^i $[\rho^a]$ corresponds to the initial condition $\rho^i(t=0) = |i\rangle\langle i|$ $[\rho^a(t=0) = |a\rangle\langle a|]$. Here $|a\rangle$ is the lowest excited quasidiscrete state (indicated by the energy interval δ in Fig. 1) which is incoherently pumped with a rate Γp_a, where p_a is the stationary occupation of $|a\rangle$ and $\Gamma = 2\pi V^2$ is the rate of escape from $|\varphi_\nu\rangle$ into the right-hand continuum. The matrix element (actually the Rabi frequency) $v(E)$ describes the optical coupling (through an electric field with amplitude E_0) between the ground state $|i\rangle$ and the Fano eigenstates $|E\rangle$ at energy E and determines the absorption [13],

$$\mathrm{Im}\chi_{\rm abs}(\omega) = (\pi/E_0^2)|v(E=\omega)|^2 .$$

The extended left-hand continuum is modeled by N quasidiscrete states $|\varphi_\nu\rangle$, forming a band with a constant density of states g which is adequate for the two-dimensional case. The stimulated emission from the incoherently pumped states at energy E_a in the interval δ can be written as

$$\mathrm{Im}\chi_{\rm em}(\omega) = (\Gamma/2E_0^2)\left|\int dE\, v(E) a(E)/(E-\omega^+)\right|^2 ,$$

with $\omega^+ = \omega + i\eta$, $\eta > 0$. As an illustration we have chosen $N = 100$, $\Delta = 1$, $\delta = 0.01$, and $g = 100$. The interval δ is then represented by the state $\langle\varphi_1| \equiv \langle a|$ $(E_1 = E_a)$;

thus $a(E)=\langle\varphi_1|E\rangle$. For this choice we have [2]

$$v(E)=-u\left[\frac{g\Delta}{N}\frac{q(E_a)}{\epsilon_1}+1\right]\left[1+\left(\frac{g\Delta}{N}\sum_{\nu=1}^{N}\frac{1}{\epsilon_\nu}\right)^2\right]^{-1/2}$$

and

$$a(E)=-\frac{1}{\pi V\epsilon_1}\left[1+\left(\frac{g\Delta}{N}\sum_{\nu=1}^{N}\frac{1}{\epsilon_\nu}\right)^2\right]^{-1/2},$$

where $\epsilon_\nu=2(E-E_\nu)/\Gamma$ and u is the Rabi frequency for the right-hand continuum alone. The well-known results [2,12] are recovered (in units of $\pi u^2/E_0^2$),

$$\mathrm{Im}\chi_{\mathrm{em}}(\omega)=\frac{\Gamma^2}{4}\frac{q^2+1}{\Delta\omega^2+\Gamma^2/4},$$

$$\mathrm{Im}\chi_{\mathrm{abs}}(\omega)=\frac{\Gamma^2}{4}\frac{(q+2\Delta\omega/\Gamma)^2}{\Delta\omega^2+\Gamma^2/4},$$

if there is only one quasidiscrete state $|a\rangle$ ($\Delta\omega=\omega-E_a$).

Figure 2(a) shows emission and absorption spectra for $q(E_a)=q_1=10$. In order to simulate the finite experimental resolution we have broadened the spectra, taking $\mathrm{Im}\omega^+=0.01$. (The unbroadened absorption spectrum consists of N zeros at the energies E_ν, $\nu>1$, and maxima in between [2].) Figure 2(b) corresponds to the situation with just one single discrete state $|\varphi_1\rangle$. Two distinct effects can be seen. In Fig. 2(a) ($N=100$), both the absorption and the emission lines are considerably narrowed as compared to the lines in Fig. 2(b) which are determined by the escape rate Γ. In fact, the linewidth in Fig. 2(a) is solely determined by the artificially introduced broadening parameter $\mathrm{Im}\omega^+=0.01$. More detailed calculations for $N\geq 100$ show that for this particular value of $\mathrm{Im}\omega^+$ the results turn out to be independent of N. The second important feature of Fig. 2(a) is that besides this interference narrowing the emission line is now much higher than the absorption line and exceeds the latter by a factor of 5.

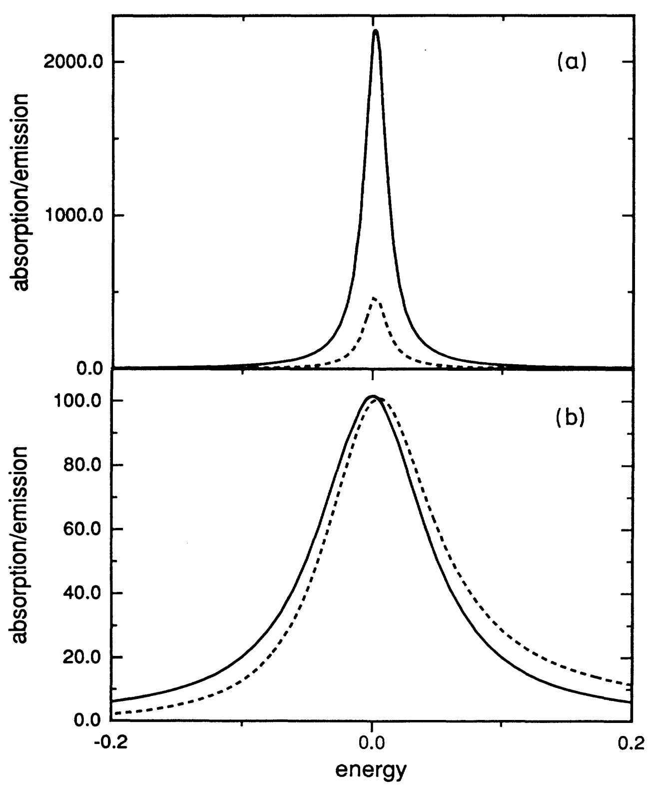

FIG. 2. Emission (solid line) and absorption (dashed line) spectra in units of $\pi u^2/E_0^2$ for (a) $N=100$ quasidiscrete levels in the interval $\Delta=[0,1]$. Only the lowest level is optically coupled to the ground state, $q_1=10$, $q_2,\ldots,q_N=0$, $\Gamma=0.1$, and $\eta=0.01$. (b) Same as (a), but the optically inactive continuum has been omitted and $\eta=0$.

The homogeneous broadening due to the escape rate Γ from a single state [Fig. 2(b)] is suppressed in the lines characteristic for coupled continua [Fig. 2(a)]. The sharpening is due to the indirect coupling of the optically inactive exciton continuum states to the lowest direct excitonic state via the electron-hole continuum, i.e., due to overlapping resonances. The absorption survives only close to the allowed strong transition as a consequence of the sum rule which applies to $|v(\omega)|^2$. (As noted by Fano [2] this sum rule is only approximately obeyed if q and Γ are taken to be energy independent.) Furthermore, the profiles do not show the typical Fano shape.

The optically inactive continuum states above the interval δ lead to a degradation of the absorption also for transitions into the states beyond the interval Δ. This degradation is even more pronounced in the results shown in Fig. 3, where we have assigned a small q value to the lowest left-hand $|i\rangle$-$|a\rangle$ transition. Now the absorption is strongly reduced relative to that of the $|i\rangle$-$|c_E\rangle$ transition, which is unity. We note, however, that the ratio of the absorption and emission strengths remains unaffected.

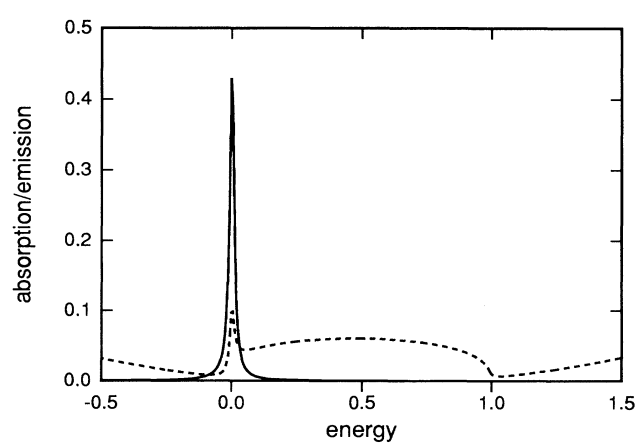

FIG. 3. Emission (solid line) and absorption (dashed line) spectra in units of $\pi u^2/E_0^2$ for the same set of parameters as in Fig. 2(a), except for $q_1=0.1$ here. The continuum background absorption is unity.

This situation actually corresponds to the conventional type-I superlattices.

In order to discuss the possible experimental demonstration of the effect predicted here we finally consider the crucial assumptions and requirements: It is fundamental that the mechanism for coupling between the direct and indirect continuum is different from the one which is responsible for the optical coupling between the ground state and the indirect continuum since otherwise no interference of the two sets of optical transition amplitudes occurs. Therefore for the experimental verification of the predicted effects, type-II structures seem to be favorite candidates as compared to bulk alloy semiconductors like AlGaAs, where disorder is responsible for both the coupling of the Γ and X states and the optical coupling of the ground state and indirect excited states. Yet we have to consider that interface roughness will cause inhomogeneous line broadening. It is clear that this inhomogeneous broadening is not affected by the Fano interference narrowing. We, therefore, expect narrow exciton lines in type-II structures only if the inhomogeneous linewidth is sufficiently small. In any case we predict that homogeneous broadening is absent in this situation, and thus the observed linewidth is caused solely by inhomogeneous broadening.

Homogeneous broadening can be inferred from dephasing experiments. The dephasing time constants for GaAs and GaAs-based type-I structures have been determined to be about 5 ps [14], corresponding to a homogeneous width of 0.13 meV (acoustic-phonon scattering). In type-II structures, however, dephasing of the direct excitons is determined by Γ-X scattering, which can be as fast as 100 fs [9], corresponding to a homogeneous width of 6.6 meV. Presently, the linewidths reported for type-II structures are dominated by inhomogeneous broadening and are typically of the order of 30 meV [15], which is still too broad to detect homogeneous linewidth narrowing. The challenge from the technological point of view is to grow samples with just the right amount of interface roughness to provide sufficient coupling of the Γ and X states without causing too large an inhomogeneous broadening.

Finally, we would like to point out that the requirement of incoherent pumping of the lowest exciton states, which is a further requirement for the observation of nonreciprocal absorption and emission line shapes, can be easily fulfilled by applying nonresonant optical excitation followed by intrawell energy relaxation as described in Ref. [9].

In conclusion, we have shown that type-II semiconductor quantum-well and superlattice structures are very promising for the observation of Fano-type interference of optical transition amplitudes. In these structures this interference will result in pronounced nonreciprocal absorption and emission profiles and interference narrowing due to overlapping resonances. Experimental demonstration seems feasible, yet careful optimization of the sample properties is required.

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