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Modification of Fano resonances by resonant polaron coupling in bulk GaAs

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Abstract. Strong modifications of the magneto-excitonic Fano lineshapes have been observed in the photoluminescence-excitation spectra of bulk GaAs when the energies of the transitions are tuned by an external magnetic field. We identify the origin of these changes as resonant polaronic coupling between the electrons or holes, forming the excitons, with longitudinal-optical phonons, in conduction and valence bands respectively. The resonant polaron effect alters the magneto-excitonic discrete states, destroying the quantum interferences responsible for the Fano asymmetry.

1. Introduction

Optical absorption investigations of bulk GaAs in a magnetic field have revealed the existence of a multiplicity of transitions between Landau levels (LLs) [1, 2]. Recently, it has been shown that the lineshapes of the magneto-excitonic transitions observed in optical absorption [3] and pseudo-absorption spectra [4] are asymmetric due to the development of Fano resonances [5]. These resonances have been reported in a great variety of experiments, such as x-ray absorption in atomic physics [6] and Raman spectroscopy in semiconductor physics [7]. In the case of magneto-optical experiments, the Fano profiles are produced by the quantum interference between the discrete excitonic states and the energetically degenerate continuum of states, which remains along the direction of the applied magnetic field. Therefore, the asymmetry of the Fano lineshape should be very sensitive to additional interactions of the discrete states, for instance polaronic coupling in polar semiconductors.

In this case, the interaction of the carriers with longitudinal-optical (LO) phonons, through the polarization field, originates a new pseudo-particle, known as polaron [8]. The polar interaction modifies the LLs in the following way [8]:

(i) they are shifted to lower energies;

(ii) the slopes of the LLs versus magnetic field are changed because of the mass renormalization of the carriers;

(iii) the LLs do not cross the virtual energy level formed by the lowest LL plus one optical phonon, leading to a splitting of degenerate energy levels (resonant polaronic coupling); and

(iv), in the limit of high magnetic field, the LLs are pinned to the energy of this virtual level.

The most common magneto-optical methods used to observe these phenomena are cyclotron resonance and interband magnetoabsorption. In the weak polar semiconductor GaAs, polaronic phenomena have been studied by cyclotron resonance [9], interband magneto-optical absorption [10], luminescence [11, 12] and Raman scattering [13]. In general, is difficult to observe the resonant polaronic coupling in the energy range of the LO phonon by infrared optical transmission experiments because polar semiconductors are basically opaque due to the reststrahlen band. Interband spectroscopy allows one to circumvent this obstacle by observing the polaronic resonance, which takes place either in the conduction or the valence band, in the pair transitions which involve those bands.

In this paper we report on the resonant polaronic coupling of the electrons and holes, forming excitons, with LO phonons and on the concomitantly induced modification of the Fano lineshapes. A related resonant coupling between electrons (and/or holes) and LO phonons has been studied in the magneto-Raman scattering in bulk GaAs [13], where double-resonances take place when the energy separation of valence LLs is tuned by the magnetic field to match the energy of LO phonon. It has also recently been shown that magneto-excitons, as opposed to single-particle states, are the particles involved in the double-Raman resonances in GaAs quantum wells [14]. This fact could be responsible for the elusiveness of resonant

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polaron coupling in the magneto-optical experiments of two-dimensional GaAs.

2. Experimental details

We have investigated a high-quality GaAs film grown by molecular beam epitaxy on a semi-insulating GaAs substrate. A 200 nm GaAs buffer layer was followed by 500 nm of $Al_{04}Ga_{0.6}As$, a 500 nm GaAs layer, a 300 nm $Al_{04}Ga_{0.6}As$ and a cap layer of 10 nm GaAs.

Photoluminescence excitation (PLE) spectra were measured at 2 K with the sample immersed in a liquid He bath cryostat within a standard-coil superconducting magnet. The magnetic field, up to 13.5 T, was applied in Faraday configuration, with $\boldsymbol{B}$ along the growth axis. The exciting light, from a Ti-sapphire laser pumped by an Ar^+ ion laser, was circularly polarized using an achromatic $\lambda/4$ plate. The luminescence was analysed by a double-grating monochromator and detected by standard photon counting techniques. For each run, the laser power was measured simultaneously with the PLE intensity in order to normalize the spectra.

The high quality of the sample was manifested by the observation of the $n = 2$ and $n = 3$ excitonic excited states, in the zero-field PLE spectra [4] and by the narrowness of the lines ($\sim$ 0.5 meV). The three-dimensional behaviour of the GaAs 500 nm layer was confirmed by the similarity of the PLE spectra taken for different tilting angles of the field with respect to the growth direction.

3. Results and discussion

Let us begin by briefly recalling the results of the theoretical description of LLs in cubic semiconductors as developed by Luttinger and Kohn [15] and later extended by Roth *et al* [16], which will be used in the analysis of the transitions observed in our PLE spectra.

In the conduction band there are two series of LLs, $|\alpha_n\rangle$ and $|\beta_n\rangle$, for spin up and spin down respectively, being n the Landau quantum number. The degenerate valence band forms four series of non-equally spaced levels $|a_n^+\rangle$, $|a_n^-\rangle$, $|b_n^+\rangle$ and $|b_n^-\rangle$, whose wavefunctions are linear combinations of the those of the hole states at zero magnetic field, $|j, m_j\rangle$, and of harmonic-oscillator wavefunctions Φ_n (see equation (7) of [11])

$$|a_n^\pm\rangle = a_1^\pm|\tfrac{3}{2}, +\tfrac{3}{2}\rangle\Phi_{n-2} + a_2^\pm|\tfrac{3}{2}, -\tfrac{1}{2}\rangle\Phi_n$$
$$|b_n^\pm\rangle = b_1^\pm|\tfrac{3}{2}, -\tfrac{3}{2}\rangle\Phi_{n-2} + b_2^\pm|\tfrac{3}{2}, +\tfrac{1}{2}\rangle\Phi_n. \qquad (1)$$

The set of corresponding eigenvalues obtained by increasing n by integer steps is known as a 'ladder' [17]. In the classical limit, the spacing between levels becomes uniform within any ladder. The plus ladders have a level spacing corresponding to 'light' holes, while the minus ladders are associated with 'heavy' holes (the character heavy or light refers to the mass in the plane perpendicular to the magnetic field direction). It is convenient to keep the levels grouped in this way when considering the selection rules for optical transitions.

In contrast to simple bands, where optical transitions are allowed only between levels with the same Landau indices, i.e. $\Delta n = 0$, the degenerate valence band of GaAs gives rise to transitions with $\Delta n = 0, -2$. The allowed interband optical transitions for right- and left-handed circular polarization of the light are [16]

$$|a_n^\pm\rangle \Leftrightarrow |\alpha_n\rangle \qquad |b_n^\pm\rangle \Leftrightarrow |\beta_n\rangle \qquad (\sigma^+)$$
$$|a_{n+2}^\pm\rangle \Leftrightarrow |\alpha_n\rangle \qquad |b_{n+2}^\pm\rangle \Leftrightarrow |\beta_n\rangle \qquad (\sigma^-) \qquad (2)$$

respectively. This selection rules will be used to identify the magneto-excitons observed in the PLE spectra.

The energy of a LL with respect to the bottom (top) of the conduction (valence) band is called the magnetic energy of the level [2], and it is given by

$$E_n^{e(h)} = (n + 1/2)\hbar\omega_c^{e(h)} \qquad (3)$$

with $\omega_c^{e(h)} = eB/m^*_{e(h)}$ the cyclotron frequency of the carrier, and $m^*_{e(h)}$ the effective mass of the electron (hole). Considering the magnetic energies of the levels both in the conduction and in the valence bands to be positive quantities, the sum of the magnetic energies of the initial and final states of a transition constitutes the magnetic energy of the transition.

Whenever the energy $E_n^{e(h)}$ of the electron or hole lies one LO phonon energy, $\hbar\omega_{LO}$, above the ground state energy, $E_0^{e(h)}$, a resonant coupling takes place between both LLs, giving rise to a splitting between the upper and lower magneto-polaron branches. The polaron thresholds, neglecting excitonic effects, have an energy given by

$$\hbar\omega_t^{e(h)} = E_g + \left(\frac{\hbar\omega_c^{e(h)}}{2} + \hbar\omega_{LO}\right)\left(1 + \frac{m^*_{e(h)}}{m^*_{h(e)}}\right) \qquad (4)$$

with E_g the energy gap of the semiconductor. Therefore, the difference between $E_n^{e(h)}$ and the ground state, at threshold, is given by

$$\Delta_t^{e(h)} = \hbar\omega_{LO}\left(1 + \frac{m^*_{e(h)}}{m^*_{h(e)}}\right). \qquad (5)$$

Figure 1 shows the PLE spectra at $B = 9$ T for σ^+ and σ^- polarization of the exciting light. The strongest transitions have been labelled according to the results of Vrehen [2]. In both polarizations the transitions are grouped in families with the same valence band Landau index. Transitions with $\Delta n = 0$ are observed in σ^+ polarization, while 'magnetically activated' transitions, $\Delta n = -2$, are resolved in the σ^- spectra. The lineshapes of the magneto-excitonic transitions between LLs in the conduction and valence bands are asymmetric when their energy is greater than that of the energy gap (marked by a double arrow in figure 1). These asymmetric structures have been identified as Fano resonances, deriving from the interaction of the magneto-excitons with the degenerate continuum of states which remains along the field direction [3, 4]. The energetically lowest magneto-excitonic transition and its excited states, which always lie at energies below any continuum of states, keep their Lorentzian lineshapes independently of the magnetic field strength. On the contrary, the

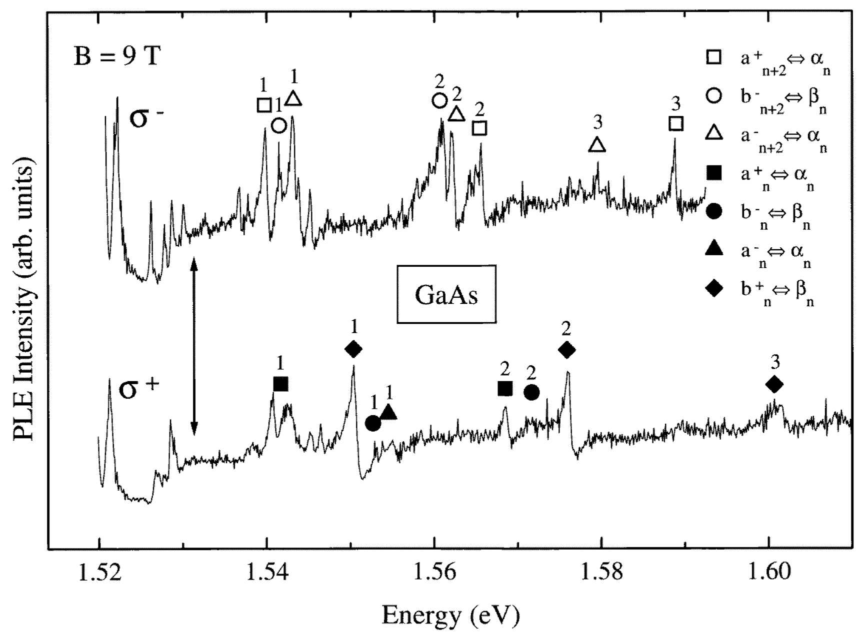

Figure 1. Low-temperature, T = 2 K, PLE spectra of bulk GaAs for σ^+ and σ^- excitation in an external magnetic field of 9 T. The transitions are identified following [2]. The double arrow indicates the continuum of the n = 0 interband transition.

magneto-excitons lying at energies larger than the bandgap are degenerate with the one-dimensional continuum, and therefore present asymmetric Fano lineshapes. The PLE profile of a Fano resonance can be written as [4, 5]

$$I \propto \frac{(q+\varepsilon)^2}{1+\varepsilon^2} \tag{6}$$

where q is a dimensionaless lineshape parameter [5], and ε is a reduced energy given by

$$\varepsilon = \frac{2(E-E_f)}{\Gamma} \tag{7}$$

with E_f and Γ the energy of the discrete transition and its broadening, modified by the coupling with the continuum states, respectively.

Figure 2 depicts the experimental Fano profile of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ transition at 6 T (full circles) together with its best fit to equation (6). Note that additional fine structure (marked by arrows) is observed in the right wing of the Fano resonance, due to the presence of other excitonic states of the same interband transition. The parameters of the fit are given in the figure.

Let us concentrate now on the $|b_n^+\rangle \Leftrightarrow |\beta_n\rangle$ Fano resonance and its evolution with magnetic field. In general, the asymmetry of the lineshapes is practically independent of the Landau index n, as can be observed for the transitions labelled 1⧫ and 2⧫ in figure 1, and also independent of the magnetic field strength. This fact is corroborated by the fits to Fano profiles, which obtain q values not very dependent on n or B. However, the Fano lineshapes are strongly modified when their energy is $\approx 2\hbar\omega_{LO}$ above the ground state exciton (see peak labelled 3⧫ in figure 1). Since the effective masses of electrons and holes, $m_e^* = 0.065m_0$ and $m_{lh}^* = 0.087m_0$, are similar [18], this energy corresponds to the threshold for the onset of resonant polaron interaction defined in equation (4).

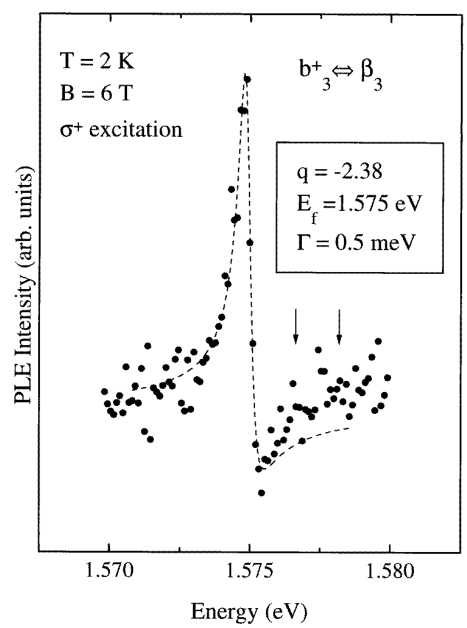

Figure 2. Low-temperature, T = 2 K, PLE spectrum (full circles) of bulk GaAs in the region of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ magneto-exciton at 6 T. The curve shows the best fit using equation (6). The arrows point to excited excitonic states of the same interband transition.

Figure 3 shows the evolution of the Fano resonance $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ with magnetic field. It is clearly seen that the resonance, asymmetric at 6 T, gradually loses its asymmetry and quenches until it completely disappears at 8.5 T. Simultaneously, a new peak, which emerges at slightly

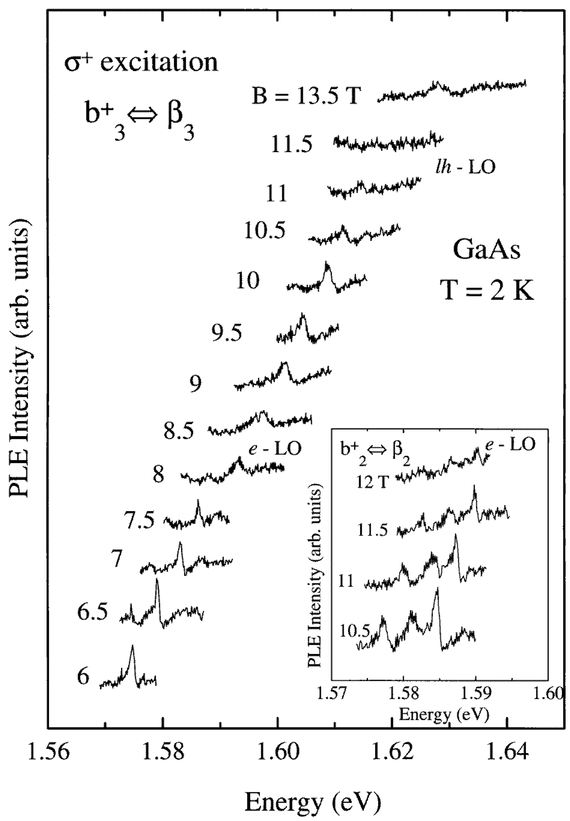

Figure 3. Evolution of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ transition with the magnetic field. The particle involved in the resonant polaron effect, electron or hole, is indicated by e–LO and lh–LO respectively. The inset depicts the evolution of the $|b_2^+\rangle \Leftrightarrow |\beta_2\rangle$ transition.

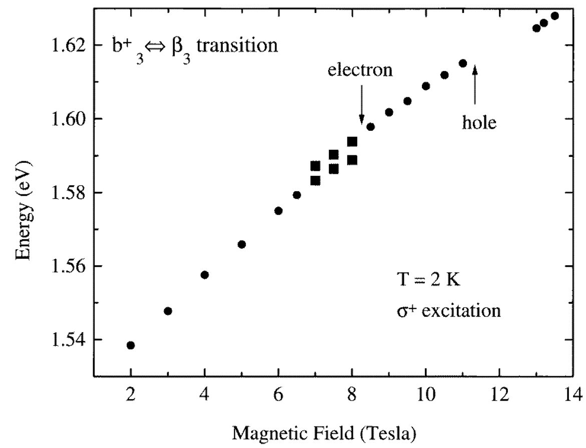

Figure 4. Energy of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ magneto-exciton as a function of the magnetic field. The squares correspond to the two branches of the magneto-polaron. The threshold fields for the resonant polaron effect are indicated by arrows.

higher energy, grows, becomes narrower and acquires an asymmetric lineshape. In a small field interval of 1 T both structures coexist and share their oscillator strength. At higher fields this Fano resonance undergoes a second quenching and vanishes at 11.5 T. We recognize again the development of a new peak at the maximum field of our magnet, 13.5 T. As we will see below, these quenchings of the resonances are due to the resonant polaron coupling of the LO phonon with the electron (e–LO) and the hole (lh–LO) respectively forming the exciton. The strong modifications of the Fano resonances are also demonstrated, in the inset of figure 3, by the evolution of the $|b_2^+\rangle \Leftrightarrow |\beta_2\rangle$ resonance, which fades away at ~ 12 T.

Figure 4 shows the energy of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ transition as a function of the magnetic field. A doublet is resolved between 7 and 8 T, whose lower branch lies ~ 64 meV above the magneto-excitonic ground state. This energy corresponds to the threshold for resonant polaron coupling of the electron, Δ_t^e, defined by equation (5), if we take $\hbar\bar{\omega}_{LO} = 36.3$ meV, $m_e^* = 0.065m_0$ and $m_{lh}^* = 0.085m_0$. The effective mass for the light holes agrees with the value of $0.087m_0$ compiled by Adachi [18]. The splitting between the two branches of the polaron amounts to ~ 3.5 meV, in reasonable agreement with theoretical calculations [19]. At the second quenching of the resonance, which takes place at ~ 11 T, the magneto-exciton lies 85 meV above the ground state. This energy difference corresponds to the threshold for resonant polaron coupling of the hole, $\Delta_t^h = 84$ meV, obtained from equation (5). An additional confirmation of the fact that the resonant polaron coupling is observed both for the electrons and holes forming the excitons is obtained from the ratio of the magnetic field strengths at which the quenchings of the Fano resonances occur, $B_h^{(3)}/B_e^{(3)} = 1.34 \pm 0.08$, which agrees with the ratio of the effective masses, $m_h^*/m_e^* = 1.31$. The mechanism responsible for the extinction of the $|b_2^+\rangle \Leftrightarrow |\beta_2\rangle$ resonance is the polaron coupling of the electron, as can be deduced from the ratio $B_e^{(3)}/B_e^{(2)} = 0.67 \approx 2/3$, which is inversely proportional to the ratio of the corresponding Landau numbers. The quenching of this resonance by the coupling of the hole will happen at higher magnetic fields than those available in our set-up.

The resonant polaron effect destroys the quantum interferences responsible for the Fano asymmetry: the peaks become broader and smaller, due to coupling with LO phonons, and lose their asymmetry. Figure 5 depicts the parameters characterizing the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ resonance, q and Γ, obtained from the fits using equation (6). At 8 T, the circles (squares) represent the values of the lower (upper) branch of the polaron. At low fields q has a value of -4, which remains constant up to ~ 7 T, then exhibits a sharp drop and has a singularity at ~ 8 T, where q has a value of -120. At this field, corresponding to the resonant polaron coupling of the electron, the lineshape is essentially symmetric. For larger fields q increases again, reaching a value of -4 at 9 T; a second dip develops at fields when the resonant polaron coupling of the hole occurs. The broadening parameter of the Fano resonance, Γ, also reflects the resonant coupling of the carriers with the LO phonon: a strong increase of the width of the resonance, triplicating its value, happens at ~ 8 T, and a new enlargement is observed above 10 T.

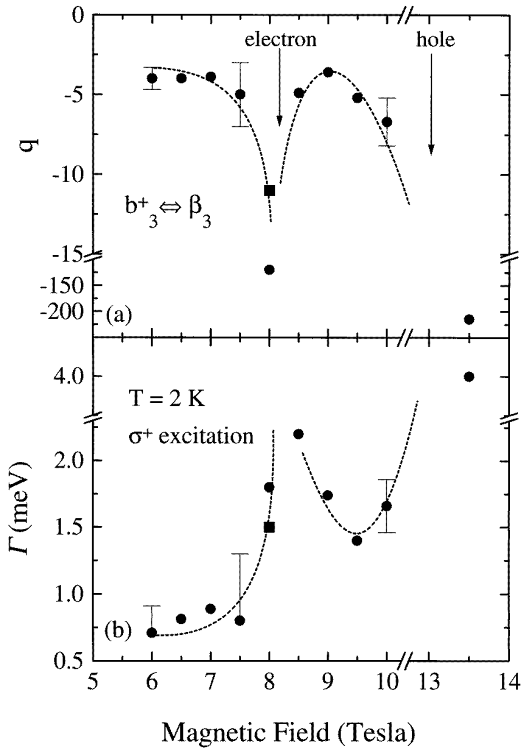

Figure 5. Lineshape parameters, q (a) and Γ (b) from the best fits using equation (6) of the $|b_3^+\rangle \Leftrightarrow |\beta_3\rangle$ magneto-exciton as a function of the magnetic field. The curves are guides to the eye. Notice the break in the scales for the values corresponding to coupling of the holes (13.5 T).

Our experiments indicate that for the two particles building the exciton, electron and hole, the resonant polaron coupling is stronger for the hole:

(i) the quenching of the Fano resonance is complete in this case, while one of the branches of the polaron remains resolvable for the coupling of the electron;

(ii) at 13.5 T, the Fano resonance is heavily broadened, $\Gamma = 4$ meV, and the lineshape is totally symmetric, $q = -210$.

Since the carrier–LO phonon interaction due to the Fröhlich mechanism is identical for electrons and holes, the stronger coupling of the hole could arise from the additional contribution of deformation-potential scattering. Moreover, the presence of the heavy-hole ladders gives extra scattering paths for the light holes, broadening the resonances. Thus, the shorter lifetime of the holes, also responsible for the lack of observation of coupled plasmon–LO modes in p-type GaAs [20], could hinder the observation of the two branches of the magneto-polaron.

4. Conclusions

We have observed a strong modification in the Fano profiles of the magneto-excitons in bulk GaAs due to resonant polaron coupling. This coupling, which occurs separately for both particles forming the exciton, destroys the coherence needed for the development of a Fano lineshape, producing striking singularities in the broadening parameter and the asymmetry of the magneto-excitons. The magneto-exciton is completely quenched when the hole is the particle involved in the coupling. From our results we obtain a value of the effective mass of the light-holes of $(0.085 \pm 0.002)m_0$.

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