Pressure-induced resonance broadening of exciton line shapes in semiconductors: Direct determination of intervalley scattering rates in GaAs

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We show that the exciton photoluminescence line shape in the GaAs/$Al_xGa_{1-x}As$ quantum wells under pressure is broadened by hybridization of the Γ exciton with the X and the L continua via electron-phonon coupling. Furthermore, we demonstrate the pressure tuning of the resonance-broadening effect which can be used to extract the electron-phonon coupling parameters directly. For GaAs we estimate the intervalley electron-phonon deformation potential $D_{\Gamma X}$ to be 10.7 ± 0.7 eV/Å. The resonance effect should be observable in other semiconductors as well.

I. INTRODUCTION

Intervalley scattering of electrons in the conduction band of a semiconductor plays an important role in high-field transport properties, in relaxation of photoexcited electrons, in processes such as light absorption in indirect semiconductors, and in a host of other phenomena such as the Gunn effect. Even though a number of factors such as the presence of impurities, composition fluctuations, or even electron-hole scattering can sometimes affect the intervalley scattering rates, in most practical situations it is primarily governed by the phonon-assisted scattering between the valleys. The electron-phonon deformation potential, which is a quantitative measure of the phonon-assisted intervalley scattering strength, is therefore a quantity of fundamental interest and has been studied for a long time both theoretically and experimentally. In this paper we determine the intervalley scattering rates using a pressure-induced Fano resonance broadening of the exciton photoluminescence line shapes in semiconductors.

The Fano resonance effect[1] is a well-known effect and has been observed in a variety of physical systems. It is the broadening of a localized level caused by resonance interaction with a continuum. The original work of Fano dealt with the double excitation of the helium atom, where the double-excited state overlaps with the spectrum of a single-excited state plus the continuum giving rise to the characteristic asymmetric line shape in the absorption spectrum. In solid-state physics this effect was first introduced by Anderson[2] in connection with *s-d* interaction in transition metals and since then it has been applied to a large number of situations. The impurity Anderson model which differs from the Fano model by an extra Coulomb term has also been applied to a variety of problems including the heavy fermion systems and the high-T_c superconductors.

Fano resonance of excitons has been recently observed in quantum wells where an exciton belonging to a higher subband interacts with the two-dimensional continuum of the lower subband either via alloy fluctuations[3,4] or via valence-band mixing.[5] Here, we describe a different type of resonance effect, where the bulklike Γ exciton interacts with the X and the L continua via electron-phonon coupling. We use this effect to extract electron-phonon coupling parameters.

II. THE RESONANCE EFFECT

Figure 1 illustrates in the bulk semiconductor the pressure-induced resonance effect of excitons, the subject of study in this paper.[6] Figure 1(a) shows the photoluminescence (PL) process from the exciton level. In an ordinary semiconductor, say, GaAs, the photoluminescence line shape is rather sharp with a linewidth of $\lesssim 3$ meV. This is caused by *intra*valley scattering of the excitons due to phonons[7] which remains more or less unchanged in the pressure range studied here. Now under applied pressure the Γ conduction minimum moves up in energy and crosses the bottom of both the L and X conduction bands, L_c and X_c, at a crossover pressure P_c of about 40 kbar.[8] The situation, illustrated in Fig. 1(b), now introduces a coupling between the Γ exciton and the X

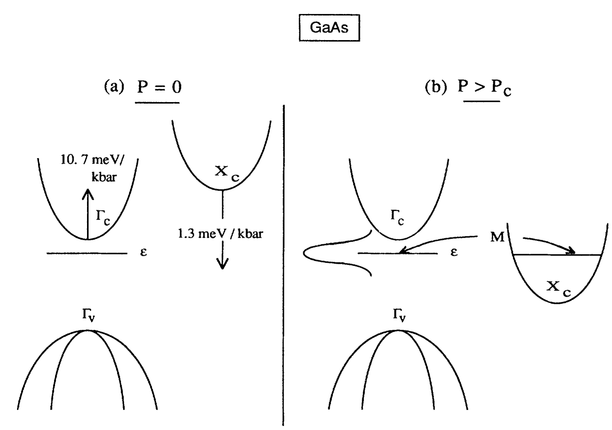

FIG. 1. Illustration of the pressure-induced resonance effect of excitons in a semiconductor. Because the energy of the electron-hole continuum relative to the exciton energy can be changed in a controlled manner by applying pressure, the resonance effect can be pressure-tuned as discussed in the text.

and the L continuum via intervalley scattering of the electron. The net result is the resonance broadening of the Γ exciton level as can be observed via photoluminescence, for instance.

Now, since the position of the exciton relative to the continuum can be changed under applied pressure, the resonance effect can be switched on and off depending on whether or not the applied pressure P exceeds the crossover pressure P_c. For the applied pressure below P_c there are no states in the X or L valley into which the Γ electron can be scattered; consequently, there is no resonance effect. Furthermore, since the electron density of states (DOS) of the X and L continua increases with energy in a free-electron-like fashion, the effective strength of the interaction and therefore the resonance broadening can be controlled by varying pressure. This provides us with a useful tool for the determination of the intervalley scattering rates between the Γ valley and the X and L valleys.

Consider the interaction (Fig. 1) between the Γ exciton and a two-particle electron-hole continuum with the electron states derived from the X valley and the hole states derived from the Γ valley. The exciton also interacts with a similar continuum with electrons derived from the L valley but this additional interaction can be generalized in a straightforward manner as done later in this paper. Let

$$c^\dagger = \sum_k B_k e^\dagger_{\Gamma k} h^\dagger_{\Gamma k} \tag{1}$$

denote the creation operator for the Γ exciton, the exciton being a superposition of the electron-hole two-particle states of zero net momentum with the electron (creation operator $e^\dagger_{\Gamma k}$) and the hole (creation operator $h^\dagger_{\Gamma k}$) derived from the Γ valley. The two-particle continuum states, which overlap with the exciton beyond a certain pressure, consist of a hole derived from the Γ valley and an electron derived from the X valley:

$$c^\dagger_{kk'} = e^\dagger_{Xk} h^\dagger_{\Gamma k'} \tag{2}$$

with the energy $\varepsilon(k,k') = \varepsilon^{\mathrm{el}}(k) - \varepsilon^{\mathrm{hole}}(k')$, where $e^\dagger_{Xk}$ describes electron states in the X valley. The coupling between the exciton and the two-particle continuum is primarily due to the phonon-assisted scattering of the electron between the Γ valley and the X valley. The Hamiltonian describing the coupling of the exciton with energy ε to the two-particle continuum with energy $\varepsilon(k,k')$ is then given by

$$H = \varepsilon c^\dagger c + \sum_{k,k'} \varepsilon(k,k') c^\dagger_{kk'} c_{kk'} + \sum_q \hbar\Omega_{\mathrm{ph}}(q) a^\dagger_q a_q + H_{e\text{-ph}} \tag{3}$$

with

$$H_{e\text{-ph}} = \sum_{k,q} M_{kq} (a^\dagger_{-q} + a_q)(e^\dagger_{X,k+q} e_{\Gamma,k} + e^\dagger_{\Gamma,k+q} e_{X,k}) \; . \tag{4}$$

Here a_q and $a^\dagger_q$ are the phonon annihilation and creation operators, Ω_{ph} is the phonon frequency and M_{kq} is the electron-phonon (e-ph) matrix element. The acoustic phonons with momentum tending to zero will not cause intervalley scattering and therefore they are not included in the Hamiltonian. The interaction results in a renormalization of the exciton energy, correct to second order in perturbation theory:

$$\bar{\varepsilon} = \varepsilon + \sum_{kk',q} \frac{|\langle c,0|H_{e\text{-ph}}|kk',q\rangle|^2}{\varepsilon - \varepsilon(k,k') - \hbar\Omega_{\mathrm{ph}}(q) + i\delta} \; . \tag{5}$$

Here $|c,0\rangle = c^\dagger|0\rangle$, i.e., it denotes the localized exciton state with no phonon while $|kk',q\rangle = c^\dagger_{kk'} a^\dagger_q |0\rangle$ denotes an electron-hole continuum state with a phonon present. The matrix element appearing in (5) is given by

$$\langle c,0|H_{e\text{-ph}}|kk',q\rangle = B_{k'} M_{kq} \delta_{k'k+q} \; . \tag{6}$$

To evaluate this matrix element we need to know the coefficients B_k, i.e., a knowledge of the exciton wave function (1) is necessary. However, it becomes particularly simple if we assume that the hole mass is much larger than the electron mass, as is the case in GaAs. Furthermore, we neglect the momentum dependence of M_{kq}, an excellent approximation since the nature of the X conduction states remains basically unchanged over the energy range of interest. We also neglect the momentum dependence of the phonon energy $\Omega_{\mathrm{ph}}(q)$ taking it equal to the phonon energy at q corresponding to the X or the L valleys. Equations (5) and (6) then lead to the expression for the renormalized exciton energy,

$$\bar{\varepsilon} = \varepsilon + \sum_k \frac{|M_{\Gamma X}|^2}{\varepsilon - \varepsilon^{\mathrm{el}}_X(k) - \hbar\Omega_{\mathrm{ph}} + i\delta} \; , \tag{7}$$

where $|M_{\Gamma X}|$ is the electron-phonon matrix element for scattering between the Γ valley and the X valley. The shift in the exciton energy given by the principal part of the second term is expected to be small except when ε is near the edge of the continuum. The imaginary part results in a broadening Δ of the exciton level:

$$\Delta = \pi |M_{\Gamma X}|^2 \rho_X(\varepsilon - \hbar\Omega_{\mathrm{ph}}) \; , \tag{8}$$

where $\rho_X(\varepsilon)$ is the one-electron DOS per spin of the X continuum. Now, if we include the effects of the L valley and furthermore, neglect the phonon energies in the DOS term in (8) [for GaAs, $\Omega^X_{\mathrm{ph}} \approx \Omega^L_{\mathrm{ph}} \approx 30$ meV (Ref. 9)], we have

$$\Delta = \pi |M_{\Gamma X}|^2 \rho_X(\varepsilon) + \pi |M_{\Gamma L}|^2 \rho_L(\varepsilon) \; . \tag{9}$$

In addition to the broadening of the exciton line, the interference effect from the continuum leads to a characteristic, asymmetric Fano line shape under certain approximations that we mention below. The effect in essence arises from the fact that when the X/L valleys are below the Γ valley, the exciton state $|c,0\rangle$ mixes with electron-hole continuum states with a phonon present $|kk',q\rangle$ and these later states also have radiative recombination matrix elements. The PL line shape is given by the Fermi golden rule

$$L(E) = \pi \sum_i |\langle i|T|0\rangle|^2 P_i \delta(E - E_i) \; , \tag{10}$$

where $|i\rangle$ is the initial state prior to photoluminescence and P_i is the probability of its occupation. T is the transition matrix for photoluminescence. Restricting to the Hilbert space with either one or no phonon, the eigenstate $|i\rangle$ is explicitly written as

$$|i\rangle = a|c,0\rangle + \sum_{k,k'} a_{k,k'}|kk',q\rangle \ , \tag{11}$$

where the expansion coefficients are denoted by a and $a_{k,k'}$. Now to obtain the occupation probability P_i one must examine the nonequilibrium process in the presence of laser excitation. If we take it to be a constant in the range of energy E_i within ~ 50 meV or so around energy of the exciton, which seems to be a reasonable approximation, then the line shape (10) may be written as a Fano line shape[1,10]

$$L(E) = \Delta / |M|^2 T_X^2 (q+\varepsilon')^2 / (1+\varepsilon'^2) \ , \tag{12a}$$

where

$$\varepsilon' = (E-\varepsilon)/\Delta \tag{12b}$$

and

$$q = (|M| T_\Gamma / T_X)/\Delta \ , \tag{12c}$$

and T_Γ (T_X) denotes the PL transition matrix elements from the Γ (X or L continuum) to the ground state. Here Δ and q are, respectively, the linewidth and the asymmetry parameters and $|M|$ is an average e-ph matrix element. Since the other parameters entering into (12c) may be assumed to be roughly constant, the asymmetry parameter should be inversely proportional to the linewidth.

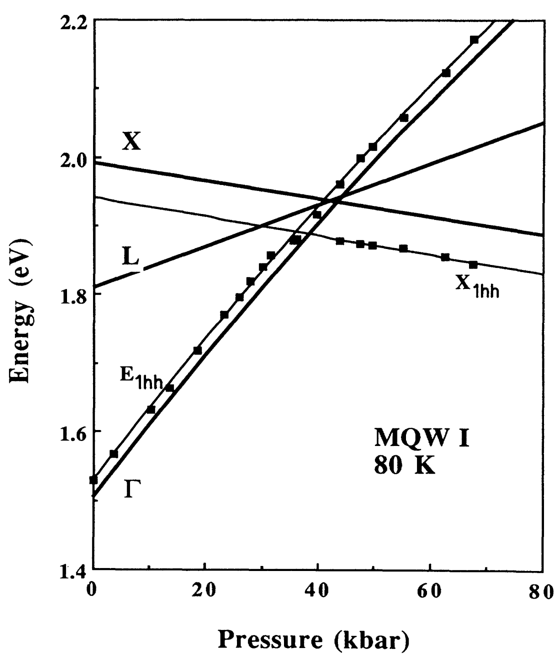

FIG. 2. Energies of the bottoms of Γ, X, and L conduction bands. The heavy lines are for bulk GaAs. The data points and the connecting solid lines are the experimental results for MQW-I. E_{1hh} represents the $n=1$ heavy-hole exciton and X_{1hh} is the staggered transition from the X conduction band in $Al_xGa_{1-x}As$ to the heavy hole in GaAs.

III. EXPERIMENT

The resonance effect discussed in this paper is applicable both for the bulk as well as for quantum wells. However, we find the quantum wells to be more suitable compared to bulk GaAs for studying the resonance effect. This is because the donor-bound exciton occurring below the free exciton dominates the PL spectrum in bulk GaAs even for relatively pure samples[11] ($\sim 10^{15}$ donors/cm^3). The bound-exciton peak loses intensity rapidly as a function of pressure beyond P_c; consequently, the resonance effect is observable only in a limited range of pressure (5–10 kbar above P_c).[12] On the contrary, the free-exciton peak which is seen in the GaAs/$Al_xGa_{1-x}As$ quantum wells is observed over a relatively large range of pressure (up to ~ 35 kbar above P_c) and therefore is ideal for

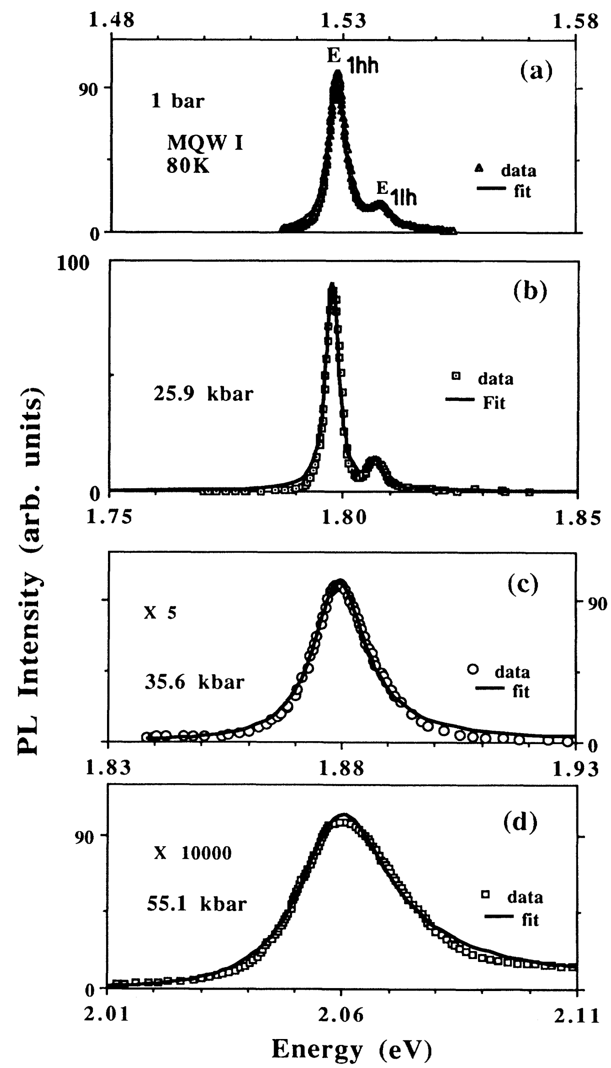

FIG. 3. The pressure dependence of the PL spectra in MQW-I. Panel (a) shows the $n=1$ heavy-hole (E_{1hh}) and light-hole (E_{1lh}) excitons which are narrow, intense, and well resolved at $P=1$ bar. The spectra are virtually unchanged in intensity and linewidth until the Γ-X crossover pressure $P_c \sim 35$ kbar as shown in (b). They progressively broaden, lose intensity, and become asymmetric for $P > P_c$ as shown in (c) and (d).

studying the resonance effect.

Two different molecular-beam-epitaxy grown quantum wells (MQW) were used in the experiments. The first (MQW-I) consists of 40 periods of 150-Å GaAs and 100-Å $Al_{0.25}Ga_{0.75}As$ layers, and the second (MQW-II) consists of the same number of periods of 260-Å GaAs and 130-Å $Al_{0.3}Ga_{0.7}As$. A gasketed diamond anvil cell was used. Pressure was monitored *in situ* via the fluorescence spectra of ruby in the pressure chamber. The pressure was hydrostatic to better than ± 1 kbar. The experimental results reported here are at $T = 80$ K. In Fig. 2 we show for MQW-I the positions of the Γ, X, and L conduction-band bottoms as a function of pressure. The data points on the Γ curve are energies of the $n=1$ heavy-hole exciton (1hh). The X data points represent the staggered transition from the X conduction band in $Al_xGa_{1-x}As$ to the $n=1$ heavy hole in GaAs. The resonance interaction, however, is much stronger within the GaAs layer since the X-Γ overlap is much weaker across the heterointerface.

Figure 3 shows the PL spectrum for several pressures for MQW-I. At $P = 1$ bar, the light-hole (1lh) and the heavy-hole (1hh) excitons are no longer degenerate in the quantum wells due to the quantum confinement effect and are clearly resolved. The linewidths are between 3 and 4 meV. The 1lh exciton occurs above the 1hh exciton with an energy separation depending on the width of the well material (~ 9 and 3 meV, respectively, for our two quantum wells). Since the 1lh exciton has the higher energy, its PL amplitude is small, the intensity in MQW-I being typically 10% of the intensity of the lower-lying 1hh exciton. In MQW-II the energy separation between the 1lh and the 1hh excitons is too small to be clearly resolved in the PL spectra.

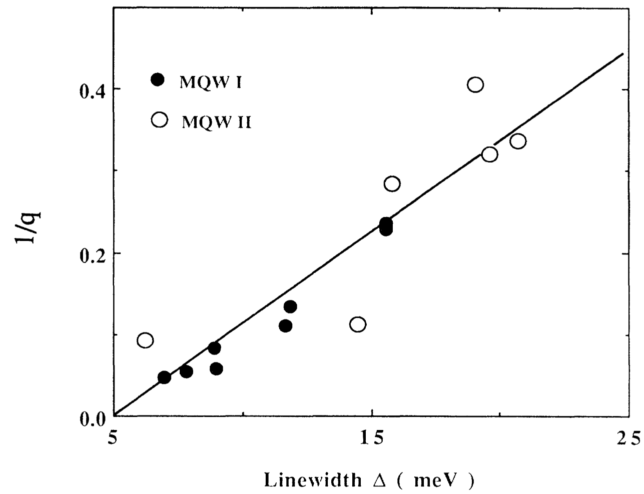

FIG. 5. The asymmetry parameter q as a function of the linewidth Δ of the PL peak beyond crossover pressure. The straight line indicates the expected behavior, Eq. (12c).

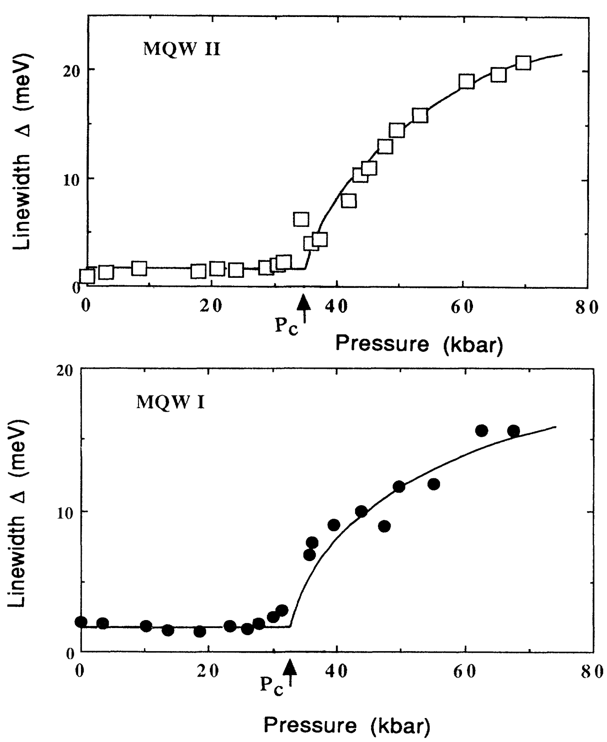

FIG. 4. Linewidth (half width at half maximum) Δ of the 1hh exciton photoluminescence peak as a function of pressure: (a) MQW-I and (b) MQW-II. Beyond P_c the exciton line shape becomes progressively broader via interaction with the continuum following roughly the parabolic $(P-P_c)^{1/2}$ dependence, Eq. (17), as indicated by the solid lines.

At higher pressures, the linewidth does not change until the Γ-X crossover pressure P_c of about 35 kbar, as shown in Fig. 2. Beyond P_c the 1hh exciton peak is progressively broadened because of the resonance effect and the 1lh peak is no longer observable. The intensity of the Γ-exciton peak is drastically reduced since electrons are now scattered to the bottom of the X band.

The PL spectra for the MQW-I are shown in Fig. 3 for four different pressures. At $P = 1$ bar the line shapes are symmetric and fit very well with two Lorentzians [Figs. 3(a) and 3(b)]. The two distinct 1lh and 1hh peaks remain virtually unchanged until the applied pressure exceeds $P_c \sim 35$ kbar. Beyond P_c the resonance broadening effect is large enough that the weaker 1lh peak is no longer resolved. The PL spectra for representative pressures just above P_c and well above it are shown in Figs. 3(c) and 3(d), respectively. The characteristic asymmetry of the line shapes is clearly seen. These line shapes fit very well with the Fano line shape, Eq. (12), from which we obtain the line-shape parameters Δ and q. The linewidth Δ as a function of pressure is shown in Fig. 4. The linewidth increases monotonically with pressure beyond P_c as the density of continuum states increases [Eq. (9)] following roughly the $(P-P_c)^{1/2}$ dependence as elaborated in the next section. The asymmetry parameter q, shown in Fig. 5, follows the expected behavior indicated from Eq. (12c), i.e., that the asymmetry parameter should be inversely proportional to linewidth Δ if we assume the parameters $|M|$, T_Γ, and T_X to be roughly constant over the pressure range of our experiment.

Now since the 1lh exciton energy lies in the 1hh exciton continuum, one might also anticipate a resonance broadening for the 1lh exciton so that the linewidth of the 1lh exciton is larger than that of the 1hh exciton as observed by Broido *et al.*[5] We do indeed find the linewidth of the 1lh exciton to be consistently broader than that of the 1hh exciton but this is a much smaller effect with the linewidth of the latter being broader by about 1 meV or so. In comparison the pressure-induced resonance broadening is a much stronger effect, with the broadening being typically in the range of 10–20 meV.

IV. DETERMINATION OF INTERVALLEY SCATTERING RATES

The PL linewidth contains direct information about the strength of the *e*-ph coupling as indicated from Eq. (9). We expect this coupling in our quantum wells to be more or less the same as that in the bulk GaAs. In terms of the *e*-ph matrix element, the deformation potential $D_{\Gamma X}$ is written as

$$|M_{\Gamma X}|=\sqrt{\hbar/(2V\rho\Omega_{\rm ph}^{X})}D_{\Gamma X}\ , \tag{13}$$

with a similar expression for $D_{\Gamma L}$. Here V is the volume of the crystal, ρ is its density, and $\Omega_{\rm ph}^{X}$ is the phonon frequency with momentum corresponding to the X minimum. Since the scattering mechanism does not induce a spin flip of the electron, in Eq. (9) $\rho_X(\varepsilon)$ and $\rho_L(\varepsilon)$ are the DOS for one spin only but include contribution from all equivalent valleys:

$$\rho_X(\varepsilon)=N_X\frac{m_X^{3/2}\sqrt{\varepsilon-\varepsilon_X}}{\sqrt{2}\pi^2\hbar_3}\ , \tag{14}$$

where ε_X is the energy of the X conduction bottom and N_X is the number of X valleys, $N_X=3$. The difference between the exciton energy and the X conduction bottom, $\varepsilon-\varepsilon_X$, varies linearly with pressure:

$$\varepsilon-\varepsilon_X=\alpha_{\Gamma X}(P-P_c)\ , \tag{15}$$

where $\alpha_{\Gamma X}$ is the pressure coefficient of the Γ conduction band relative to the X minimum. The expression for $\rho_L(\varepsilon)$ is similar to Eqs. (14) and (15) with the number of L valleys $N_L=4$. From Eqs. (9), (14), and (15) we find, neglecting the slight difference between the crossover pressures for the X and the L conduction bands, that the linewidth Δ should be proportional to $(P-P_c)^{1/2}$:

$$\Delta=(2\sqrt{2}\pi\rho\hbar)^{-1}\left[\frac{3m_X^{3/2}\alpha_{\Gamma X}^{1/2}}{\hbar\Omega_{\rm ph}^{X}}D_{\Gamma X}^2+\frac{4m_L^{3/2}\alpha_{\Gamma L}^{1/2}}{\hbar\Omega_{\rm ph}^{L}}D_{\Gamma L}^2\right]\sqrt{P-P_c}\ . \tag{16}$$

The various parameters appearing in (16) are well known except the deformation potentials which can be determined from the measured values of Δ. Taking $m_X=0.41m_e$, $m_L=0.22m_e$, the crystalline density $\rho=6065.6m_e/\text{Å}^3$, the pressure coefficients[8,13,14] $\alpha_{\Gamma X}=12$ meV/kbar and $\alpha_{\Gamma L}=6$ meV/kbar, we find

$$\Delta=0.0193(D_{\Gamma X}^2+0.37D_{\Gamma L}^2)\sqrt{P-P_c}\ , \tag{17}$$

where pressures are expressed in kbars, the deformation potentials in eV/Å, and the linewidth in meV.

The expected square-root dependence (17) is followed within experimental error by the measured linewidths shown in Fig. 4. From the coefficient of this dependence, we obtain the value of $D_{\Gamma X}^2+0.37D_{\Gamma L}^2$ to be 130 $(\text{eV/Å})^2\pm15\%$ for the two quantum wells. The experimental values reported earlier for the two deformation potentials, $D_{\Gamma X}$ and $D_{\Gamma L}$, vary between 1 and 10 eV/Å,[15] while calculations show[16,17] them to be approximately equal and in the range of 2.9–3.4 eV/Å. We cannot determine $D_{\Gamma X}$ and $D_{\Gamma L}$ individually from our measurements but rather the combination of these two. If we take $D_{\Gamma L}=6.5\pm1.5$ eV/Å as reported by Shah *et al.*[18] and Kim and Yu[19] we obtain the value $D_{\Gamma X}=10.7\pm0.7$ eV/Å.

Recently Goni *et al.*[20] studied the exciton line broadening under pressure via optical absorption in GaAs. They obtained the exciton linewidth by fitting the leading edge of the absorption spectrum to a Lorentzian convolution function. Their value of $D_{\Gamma X}=4.8$ eV/Å is low compared to our value. We do not understand the reason for this discrepancy. However, we note that values obtained by other authors[19] are similar to our value of $D_{\Gamma X}$. Finally, we note here that while in the picosecond luminescence experiments such as reported in Ref. 18, the *e*-ph deformation potential enters rather indirectly and is obtained by comparing results of a Monte Carlo calculation with the observed relaxation of the photoexcited carriers, the resonance effect described here is a more direct method for measuring the deformation potential.

V. CONCLUSION

In conclusion, we have observed the resonance broadening of excitons in the semiconductor quantum wells under pressure. The resonance linewidth Δ is found to be proportional to the DOS in the continuum band, $\rho(\varepsilon)$, corresponding to the exciton energy. We have furthermore demonstrated this resonance effect to provide a direct method for the determination of the deformation potential. For GaAs, we obtain the value for the intervalley deformation potential to be $D_{\Gamma X}=10.7\pm0.7$ eV/Å.

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