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Hole-quasiparticle resonant polaron coupling in quantum wells containing high densities of free carriers

P. E. Simmonds

Department of Physics, University of Wollongong, Wollongong, NSW 2500, Australia

M. S. Skolnick

Department of Physics, University of Sheffield, Sheffield S3 7RH, United Kingdom

T. A. Fisher

Department of Physics, University of Wollongong, Wollongong, NSW 2500, Australia

K. J. Nash

Royal Signals and Radar Establishment, St. Andrew's Road, Malvern, Worcestershire, WR14 3PS, United Kingdom

R. S. Smith

GEC Hirst Research Centre, Wembley, Middlesex, HA9 7PP, United Kingdom (Received 10 October 1991)

Strong resonant polaron coupling phenomena in the magnetophotoluminescence (magneto-PL) spectra of modulation-doped quantum wells are reported. The polaron interactions arise between hole quasiparticles in otherwise filled electron Landau levels, and LO phonons of the quantum-well material. This type of many-body polaron coupling phenomenon is expected to be a universal feature of the magneto-PL spectra of quantum wells with electron Fermi energy greater than the LO-phonon energy of the well material.

New resonant polaron coupling (RPC) phenomena in the magneto-optical spectra of quantum wells (QW's) containing high densities of free carriers are reported. The phenomena have a many-body character, since the resonances arise from the Fröhlich interaction between hole quasiparticles in otherwise filled electron Landau levels (LL's) and the LO-phonon modes of the QW material. It is shown that a common requirement for the observation of RPC in the magnetophotoluminescence (magneto-PL) of QW's is that the carrier density should be of sufficient magnitude that the electron Fermi energy is of the order of, or greater than, the LO-phonon energy of the OW material.

RPC has been much studied in both three-dimensional^{1,2} (3D) and quasi-2D (Refs. 3-6) systems, principally in cyclotron resonance (CR) experiments.⁷ In such cases, the RPC is usually observed as a strong cyclotron mass enhancement, the result of electron-LO-phonon interactions which lead to pronounced anticrossing between unoccupied LL's (LL index $N_e > 0$) and the $N_e = 0$ plus one-phonon state.^{1,8-10} In the present work, the resonant coupling is observed as an interaction between LO-phonon satellites of recombination involving electrons in upper LL states (LL index $N_e > 2$) and PL transitions from the $N_e = 0$ LL.¹¹ Large values for the resonant polaron interaction strength, in comparison with CR experiments, are observed. This is attributed to the absence of LL occupation effects⁸⁻¹⁰ for the hole quasiparticles.

The magneto-PL experiments were carried out on a series of asymmetric, modulation-doped $Al_yGa_{1-y}As$ -In_xGa_{1-x}As-GaAs ($y \approx 0.23$, $x \approx 0.1$) strained-layer

QW's with high n_s values ranging from $(0.8-1.6) \times 10^{12}$ cm⁻² and well widths of 150-250 Å. Zero-field results are discussed elsewhere.^{12,13} The particular sample investigated in detail has a QW width of 150 Å and $n_s = 1.6 \times 10^{12}$ cm⁻² ($E_F = 56$ meV), determined from Shubnikov-de Haas measurements. Magneto-PL measurements were carried out at 4.2 K in magnetic fields (*B*) up to 10 T.

The PL spectrum at B=0 is shown in Fig. 1(a). It is composed of two bands, E_{11} and E_{21} , arising from recombination of electrons in the n=1 and n=2 subbands, respectively, with photocreated holes thermalized in the lowest heavy-hole subband close to k=0. The spectra extend from the bottom of the n=1 subband at E_1 up to an energy corresponding to E_F . The $\Delta k_{\perp}=0$ in-plane wavevector selection rule is broken by disorder (or hole localization) effects^{14,15} giving rise to the E_{11} transitions, observed with strongly decreasing strength above the n=1subband edge.

 E_{21} transitions are seen since the E_F of 56 (±1) meV is very close to the $E_2 - E_1$ subband separation (56±1 meV), with an upper limit to the n=2 population of 5×10^{10} cm⁻² deduced from the absence of any LL splitting of the E_{21} PL. The E_{21} peak is ~3 times stronger than E_{11} principally because of the larger overlap of E_2 electrons with the photocreated holes than that for E_1 electrons.¹³

In magnetic field the E_{11} PL breaks up into a series of LL peaks, labeled $(N_e,0)_1$, as shown in Figs. 1(b)-1(e), arising from recombination of electrons in LL's (index N_e) with holes in the lowest valence-band LL. The

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FIG. 1. PL spectra as a function of magnetic field. (a) n=1 and n=2 subband E_{11} and E_{21} recombination is observed at B=0. In (b)-(e) splitting into LLs $(N_e,0)_1$ is observed. LO-phonon satellites N_L of $(N_e,0)_1$ are observed with resonant enhancement of intensity as they approach $(0,0)_1$.

 $\Delta N = 0$ selection rule is broken by disorder for the same reason that nonvertical transitions occur at zero field.¹⁴ The energy spacings of the higher LL transitions ($N_e > 1$) are given by the electron cyclotron energy $\hbar \omega_c$, where $\omega_c = eB/m_e$ with $m_e = 0.069m_0$.¹⁵ The E_{21} peak shows strong oscillations in intensity with field [see Figs. 1(b)-1(e)], of very similar form to those reported by Chen and co-workers.^{16,17} The interpretation of these oscillations will be discussed in detail elsewhere.

For the present paper, the most important information is contained in the low-energy peaks labeled $N_L = I_L$, 2_L , 3_L ,.... These N_L satellites arise very close to the GaAs LO-phonon energy of $\hbar \omega_{LO} = 36.7$ meV below the corresponding $(N_e, 0)_1$ transitions. The energy of the GaAslike mode of the QW, which contains only 9% In, will correspond closely to this value. The PL transition energies are plotted as a function of field in Fig. 2. The $(N_e, 0)_1$ Landau fan is reproduced by the N_L LO-phonon satellite



FIG. 2. PL energies of transitions in Fig. 1 as a function of magnetic field. The N_L transitions are one LO-phonon energy below the $(N_e,0)_1$ peaks. The resonant coupling between the 3_L and 4_L satellites and $(0,0)_1$ is clearly visible. Triangles denote E_{21} transitions and solid circles denote $(N_e,0)_1$, N_L transitions. Open symbols denote weak transitions. No features at E_{21} LO are observed.

lines displaced to lower energy by $\hbar\omega_{\rm LO}$. Very marked resonant anticrossing between the lower-lying $(N_e, 0)_1$ transitions and the N_L lines is observed around fields of 4.4, 5.5, and 7.3 T given by the "magnetophonon" resonance condition $\Delta N_e \hbar \omega_c = \hbar \omega_{\rm LO}$.¹⁸ ΔN_e is the difference in Landau index between the parent LL of the N_L satellite and the N_e level with which resonance occurs.

In the region of resonance, clear exchange of oscillator strength occurs between N_L and $(N_e, 0)_1$, as seen in Figs. 1(b)-1(e). Away from resonance the satellite 3_L for example, has < 1% of the $(0,0)_1$ intensity. As 3_L approaches $(0,0)_1$ from lower energy its intensity increases until at resonance where the two transitions are at their minimum separation $[\Delta E = 4.4 \text{ meV for } 3_L \text{ with } (0,0)_1]$ the intensities are approximately equal [Figs. 1(c) and 4]. Beyond this field the lower component is pinned to the energy of the unperturbed $(0,0)_1$ transition while the 3_L character is transferred to the upper component and its intensity decreases. Near resonance, the N_L satellite intensities far exceed those of the weak parent $(N_e, 0)_1$ zerophonon transitions. The pinning behavior and exchange of oscillator strength are characteristic of hybridizing states in general and specifically of the coupled LL-LOphonon states involved in RPC.³

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Almost identical resonance behavior is found in a similar sample which has slightly lower n_s (1.45×10¹² cm⁻², E_F =48 meV) and only one occupied electron subband. Two further asymmetric QW's of widths 200 and 250 Å with E_F between 35 and 40 meV also gave qualitatively similar resonances. In addition, strong resonant enhancement of LO satellites has been seen in an (In,Ga)As-InP QW ($n_s = 10^{12}$ cm⁻², width 100 Å).¹⁹

Unlike conventional CR measurements, in the present experiments both the parent LL of the N_L satellite and the N_e level involved in the resonance are, generally, fully occupied. The resonant coupling occurs in the final states of the PL transitions between quasiholes (unoccupied electron states) which occur in the otherwise filled electron LL's as a result of recombination. The interacting states are shown schematically in Fig. 3 and consist of (i) one "hole" in the $N_e = 0$ LL of the Fermi sea and (ii) one "hole" in the $N_e > 1$ LL, plus one LO phonon emitted in the PL process. Here $N_e > 1$ is the parent LL of the resonating N_L phonon satellite. The two states are coupled together by the Fröhlich hole-LO-phonon interaction. The resonance condition $\Delta N_e \hbar \omega_c = \hbar \omega_{\rm LO}$ will be fulfilled in the magneto-PL of all systems where the electron Fermi energy E_F is $\gtrsim \hbar \omega_{\rm LO}$ such that the energy separation between *filled* LL's can be tuned through the energy region of $\hbar \omega_{10}$.

In Fig. 4, the normalized relative intensities of the N_L satellites $[N_L$ intensity divided by the N_L plus $(0,0)_1$ intensities] are plotted against the estimated unperturbed transition energy separations. The relative N_L intensities of Fig. 4 are obtained from the observed peak intensities, after subtraction of the $(0,0)_1$ contribution, assuming constant PL linewidths with field. The accuracy is limited to approximately $\pm 30\%$ in the relative intensities, particularly close to resonance because of the difficulty in separating the contribution from two closely overlapping lines.

Close to resonance, the satellite intensities are described quite well in terms of the hybridized $(0,0)_1$ and N_L final



FIG. 3. Schematic diagram showing the resonating quasihole energy levels involved in the final states of the PL transitions. The final states without and with emission of an LO phonon are shown on the left- and right-hand sides of the figure, respectively.



FIG. 4. Plot of N_L satellite intensity divided by sum of N_L and $(0,0)_1$ intensities as a function of unperturbed energy separation between the two transitions. The dashed curve is a fit to a model of two interacting states with mixing potential $\Delta E/2 = 2.2$ meV.

states, treated as a two-level coupled system. This is a marked simplification of RPC theory,^{8,20,21} but describes behavior similar to that predicted by perturbation theory close to resonance.^{8,20} Using the value $\Delta E/2 = 2.2$ meV (from Fig. 2) for the electron-phonon mixing potential, the variation of intensities with energy separation, shown by the dashed curve in Fig. 4, is obtained. This provides a reasonable fit to the experimental points in Fig. 4, and demonstrates the expected exchange of oscillator strength between two interacting levels which anticross at resonance.²²

It is instructive to draw comparisons of the LL-LOphonon coupling phenomena observed in magneto-PL with previous studies in CR.²⁻⁶ In pure 2D, the electron-LO-phonon interaction is expected to be enhanced relative to 3D, but due to the nonzero spatial extent of the electron wave function normal to the layers and to screening, the electron-phonon coupling strength in real doped heterojunctions (HJ's) (and QW's) is reduced to values close to or less than those observed in 3D.^{8-10,20} The most detailed studies of resonant polaron effects in GaAs- $Ga_{1-x}Al_{x}As$ HJ's have been carried out by Langerak et al.⁵ These workers studied the cyclotron mass enhancements close to resonance for n_s from 0.8 to 5.4×10¹¹ cm^{-2} . The values for CR mass enhancements can be expressed in terms of the splitting (ΔE), assumed symmetric, between the upper and lower polaron branches at resonance. A very strong reduction of ΔE from ~5.0 meV at 3.4×10^{11} cm⁻², to about a factor of 2 smaller at 5.4×10¹¹ cm⁻², was observed in Ref. 5. The value of ΔE at 3.4×10¹¹ cm⁻² is close to that found at 4×10¹¹ cm⁻² by Sigg, Wyder, and Perenboom.⁶ At still higher densities $(9 \times 10^{11} \text{ cm}^{-2})$, Ziesmann, Heitmann, and Chang found no evidence for polaron coupling at resonance from CR in a 200-Å InAs QW.⁴

These results for the magnitude of the RPC in CR with increasing n_s are in strong contrast to the present PL results, where $\Delta E = 4.4$ meV is observed for the $3_L - (0,0)_1$

resonance, at the *much higher* density of 1.6×10^{12} cm⁻². The contrast between magneto-PL (resonance with $N_e = 3$ LL) and CR (resonance with $N_e = 1$ LL) is even greater when account is taken of the likely scaling of ΔE with Landau index. ΔE has been calculated to vary as $N_e^{-2/3}$ (Ref. 21) leading to predicted ΔE values $3^{2/3} \approx 2$ times *smaller* for magneto-PL in the present experiment (resonance with $N_e = 3$ LL) than for CR (resonance with $N_e = 1$ LL).

The very large strength of the resonant polaron interaction for the hole quasiparticles in PL, by comparison with CR, can be understood qualitatively by consideration of the role of LL occupation effects.^{5,8-10} In CR such occupation effects are very important. The resonant coupling arises between the $N_e = 1$ LL and the $N_e = 0$, 1-LOphonon state. As the $N_e = 0$ population increases with n_s (in Ref. 5, LL filling v=0.67 at 3.4×10^{11} cm⁻², v=1 at 5.4×10^{11} cm⁻² at resonance), the polaron interaction will be increasingly quenched, due to the occupation of the necessary scattering states in $N_e = 0$. At complete filling, the resonant contribution to the CR mass enhancement is absent.¹⁰ Good agreement has been found in Ref. 5 with calculations in which the occupation effect is the dominant many-particle factor.

By contrast in magneto-PL such occupation effects are not important, since the scattering of the holes occurs between otherwise filled LL's; all states within the resonating levels are available for the quasihole scattering since the hole levels are unoccupied. Coulomb screening alone acts to reduce the strength of the polaron interactions.²³ Extrapolation of the predictions for ΔE for a 100-Å QW in Ref. 10 to $n_s = 1.6 \times 10^{12}$ cm⁻², including only the n_s dependence of the screening (no occupation effects), leads to reasonable agreement²⁴ with the value of $\Delta E = 4.4$ meV observed in magneto-PL. However, a more detailed analysis of the present polaron phenomena clearly requires a theoretical treatment specifically directed towards the present case of hole-quasiparticle-LO-phonon coupling.

In conclusion, new resonant polaron coupling phenomena have been observed in the PL spectra of filled Landau levels in modulation-doped QW's. The results are interpreted in terms of hole-LO-phonon coupling in the final state of the Fermi sea. Such hole-quasiparticle-LOphonon coupling is expected to be a universal feature of the magneto-PL spectra of all QW's with electron Fermi energy equal to or greater than $\hbar \omega_{LO}$.

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- ²²Such an exchange of oscillator strength at resonance, of course, is characteristic of any two-level coupled system, independent of the nature of the mixing potential. The good fit to the experimental results of Fig. 4 does not imply support for any specific model for the hole-LO-phonon coupling.
- ²³Reference 10 shows that the reduction of ΔE by occupation effects is considerably larger than that due to Coulomb screening, for partially filled LL's.
- ²⁴We estimate a value of $\Delta E = 3.8 \text{ meV}$ at $n_s = 1.6 \times 10^{12} \text{ cm}^{-2}$ from the results of Ref. 10 with no occupation effects, after making allowance for the N_e dependence of ΔE of $\Delta E \propto N_e^{-2/3}$ (Ref. 21). In the absence of screening, the corresponding value would be $\Delta E \approx 5.7 \text{ meV}$ (Ref. 10).