Collective and single-particle intersubband excitations in narrow quantum wells selected by infrared absorption and resonant Raman scattering

Takeya Unuma,[1,2,*] Kensuke Kobayashi,[1,†] Aishi Yamamoto,[3] Masahiro Yoshita,[1] Kazuhiko Hirakawa,[2] Yoshiaki Hashimoto,[1] Shingo Katsumoto,[1] Yasuhiro Iye,[1] Yoshihiko Kanemitsu,[3,†] and Hidefumi Akiyama[1]

[1]*Institute for Solid State Physics, University of Tokyo, and CREST, JST, Kashiwa, Chiba 277-8581, Japan*

[2]*Institute of Industrial Science, University of Tokyo, Meguro-ku, Tokyo 153-8505, Japan*

[3]*Graduate School of Materials Science, Nara Institute of Science and Technology, Ikoma, Nara 630-0192, Japan*

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We demonstrated a direct comparison between infrared absorption and resonant Raman scattering by intersubband electronic excitations in GaAs/AlAs single quantum wells (QWs) over a wide range of subband separation 50–140 meV, or well width 10–18 nm. The two probes showed stark contrast for a narrow (10 nm) QW: Raman spectra had only one peak of single-particle excitations, while absorption spectra had only one peak of collective charge-density excitation with a remarkable energy shift due to dynamical many-body effects. The observed Coulomb interaction energies were in reasonable agreement with calculations based on the local-density functional theory.

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Infrared absorption and resonant Raman scattering are the most familiar probes for intersubband electronic excitations in doped semiconductor quantum wells (QWs), which have been used to investigate subband structures, collective modes, and many-body Coulomb interaction effects.[1,2] Intensive studies have shown that infrared absorption measures intersubband collective charge-density excitation [charge-density wave (CDW)].[3]. On the other hand, resonant Raman scattering is known to detect intersubband collective charge-density excitation for a polarization parallel to the incident-light one, collective spin-density excitation [spin-density wave (SDW)] for a crossed polarization, and single-particle excitations for both polarizations. [4]

In most cases, however, the two probes have been applied to different regimes of intersubband transition energy E_{10}, namely, infrared absorption to $E_{10} \gtrsim 100$ meV[5] and resonant Raman scattering to $E_{10} \lesssim 50$ meV.[6] Thus, direct comparison between spectra measured by the two independent probes has never been accomplished, though there have been a few trials[7,8] and theoretical suggestions.[1,2]

We have recently extended intersubband Raman measurement to GaAs/AlAs single QWs with $E_{10} \gtrsim 50$ meV and observed a change in Raman spectra.[9] The results showed that the peaks of the collective charge-density and spin-density excitations became smaller in narrower QWs and only the peaks of the single-particle excitations were found for large $E_{10} \gtrsim 100$ meV. Since infrared absorption occurs solely for collective charge-density excitation, intriguing difference between infrared absorption and resonant Raman scattering should appear in narrow QWs.

In this paper, we demonstrate a direct comparison between spectra of infrared absorption and resonant Raman scattering by the lowest intersubband electronic excitations ($E_0 \rightarrow E_1$) in identical samples of modulation-doped GaAs/AlAs single QWs. For a narrow QW with 10 nm width, spectra of infrared absorption and resonant Raman scattering are found to have only one peak with a remarkable energy difference of 8.0 meV, which is consistent with the assignments of the absorption peak and the Raman peak to collective charge-density excitation and single-particle excitation, respectively. For a wider QW with 13.5 nm width, we confirm that only one absorption peak of collective charge-density excitation is displaced from a dominant polarization-independent Raman peak of single-particle excitations, and that it is exactly at the energy of a small Raman peak of collective charge-density excitation for the parallel polarizations. Energy differences between the peaks of collective charge-density excitation and single-particle excitation indicate dynamical many-body Coulomb interaction energies, which indeed agree with calculations based on the local-density functional theory for QWs with 10–18 nm width.

The samples used in this study were GaAs/AlAs single QWs with well widths of 10, 13.5, and 18 nm grown by molecular beam epitaxy on (001) GaAs substrates.[9] The structure of the 10 nm QW sample consisted of a Si-doped $Al_{0.33}Ga_{0.67}As$ layer, a 4.0 nm undoped $Al_{0.33}Ga_{0.67}As$ spacer layer, a 6.0 nm undoped AlAs barrier, a 10 nm undoped GaAs QW, a 6.0 nm undoped AlAs barrier, and a Si-doped $Al_{0.33}Ga_{0.67}As$ layer. The other two samples had similar structures except for the well width. The central parts of wafers grown with structural parameter uniformity within ±0.7% were cut into smaller pieces for different measurements. We confirmed sheet electron concentrations N_S by measuring Shubnikov–de Haas oscillations at 4.2 K, which are shown in Table I with Hall mobilities μ.

Intersubband infrared absorption spectra were measured at 12 K with a Fourier transform infrared spectrometer and microscope. The sample pieces were processed into 3-mm-long multipass waveguide structures with 45°-polished parallel edges, and aluminum (~100 nm thick) was evaporated onto the top surfaces to serve as a gate for controlling electron concentrations in QWs. We obtained reference transmission spectra by depleting electrons.

Intersubband electronic Raman spectra were measured also at 12 K in a backscattering geometry normal to the QW layers, with incident- and scattered-light polarizations parallel (∥) or crossed (⊥). The incident photon energy E_{inc} was tuned for maximum Raman scattering intensities and reso-

TABLE I. Main parameters of the samples and dynamical many-body effects on intersubband excitation energies. N_S is the sheet electron concentration and μ is the mobility at 4.2 K. αN_S and βN_S are the depolarization shift and excitonic shift, respectively.

QW width (nm)	10	13.5	18
N_S (cm^{-2})	1.2×10^{12}	1.1×10^{12}	6.3×10^{11}
μ ($cm^2/V\,s$)	4.3×10^{4}	5.6×10^{4}	4.4×10^{5}
		Calculation	
αN_S (meV)	12.1	15.0	11.0
βN_S (meV)	3.5	3.1	2.4
β/α	0.29	0.21	0.22
$(\alpha-\beta)N_S$ (meV)	8.6	11.9	8.6
		Experiment	
$(\alpha-\beta)N_S$ (meV)	8.0	11.5	8.0

nant with an interband energy gap, which we assigned to the one between the electron first excited (E_1) subband and the heavy-hole second excited (H_2) subband in QWs from energy level calculations. Although raw original data on Raman signals for various incident photon energies, as well as experimental details, were already presented in Ref. 9, they included rather large backgrounds due to photoluminescence. In the present study, we further measured corresponding spectra under off-resonance-excitation conditions to evaluate the backgrounds, which have been subtracted in the final results.

Figure 1 shows spectra of intersubband (a) infrared absorption and (b) electronic Raman scattering of the 10 nm QW. Only one spectral peak for infrared absorption was found at 142.2 meV. On the basis of established microscopic description[1,3] of intersubband infrared absorption processes, the peak was interpreted as collective charge-density excitation. The Raman spectra for the parallel and crossed polarizations had only one peak at almost the same energies of 134.2 and 134.0 meV, respectively. Since the observed Raman peaks had almost no polarization dependence, we assigned them to single-particle excitations (SPE). No polarization-dependent Raman signal due to collective excitations was observed. We obtained similar results also for other QWs narrower than 10 nm.[9] A remarkable energy difference of 8.0 meV between the absorption and Raman peaks can be seen in Fig. 1, which will be discussed in later paragraphs.

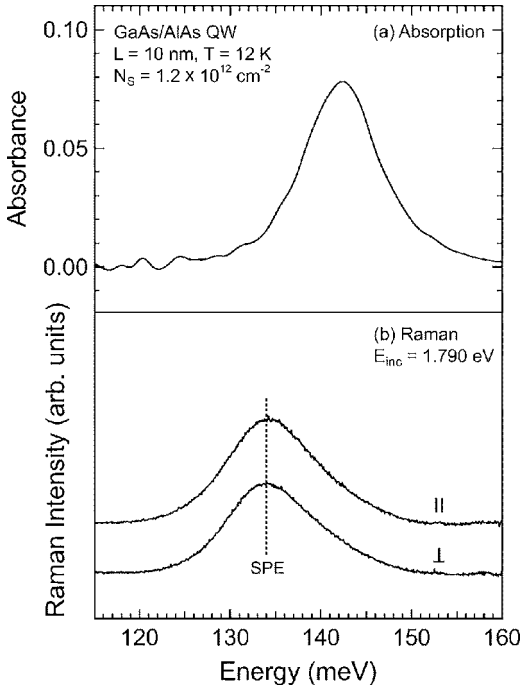

FIG. 1. Intersubband (a) infrared absorption and (b) electronic Raman spectra of a 10 nm GaAs/AlAs single QW at 12 K. The ∥ and ⊥ marks mean that incident- and scattered-light polarizations were parallel and crossed, respectively. The backgrounds due to photoluminescence in the Raman spectra have been subtracted.

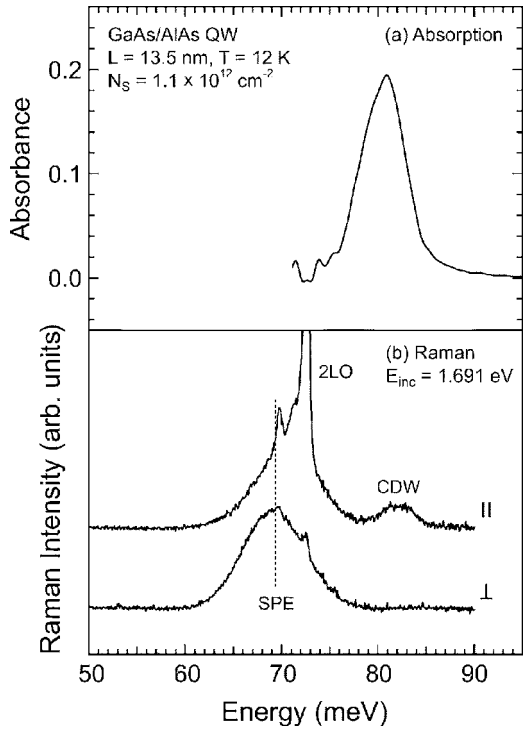

FIG. 2. Intersubband (a) infrared absorption and (b) electronic Raman spectra of a 13.5 nm GaAs/AlAs single QW at 12 K. The ∥ and ⊥ marks mean that incident- and scattered-light polarizations were parallel and crossed, respectively. The backgrounds due to photoluminescence in the Raman spectra have been subtracted.

Figure 2 shows spectra of intersubband (a) infrared absorption and (b) electronic Raman scattering of the 13.5 nm QW. Only one spectral peak for infrared absorption was found at 81.0 meV with a 5.4 meV linewidth, which was ascribed to collective charge-density excitation. The Raman spectra had several peaks. Sharp peaks at 72.6 and 69.8 meV for the parallel polarizations are from two-LO-phonon excitations in the GaAs and $Al_{0.33}Ga_{0.67}As$ layers, respectively, and should be neglected in our analysis of intersubband excitation signals. A major peak in the Raman spectra was found at 69.5 meV for both polarizations, and a small peak was found at 81.8 meV with a 3.6 meV linewidth for the parallel polarizations. From the polarization dependence,

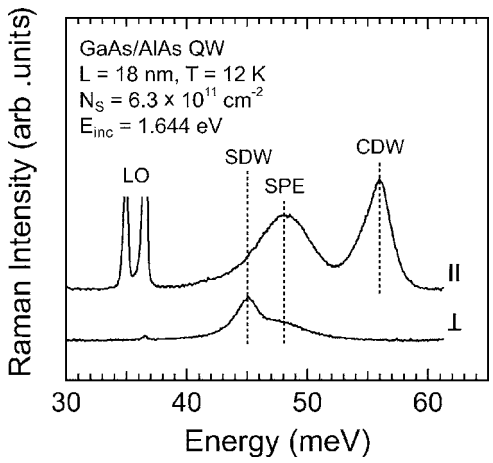

FIG. 3. Intersubband electronic Raman spectra of an 18 nm GaAs/AlAs single QW at 12 K. The ∥ and ⊥ marks mean that incident- and scattered-light polarizations were parallel and crossed, respectively. The backgrounds due to photoluminescence have been subtracted.

these peaks were assigned to single-particle excitations and collective charge-density excitation, respectively, labeled SPE and CDW. For the crossed polarizations, the peak of collective spin-density excitation was not resolved, possibly because it was overlapped by the larger peak of single-particle excitation. Comparison of these spectra has shown that the absorption peak assigned to collective charge-density excitation is clearly displaced from the major Raman peak of single-particle excitations, and that it is exactly at the energy of the small Raman peak of collective charge-density excitation.

Figure 3 shows intersubband electronic Raman spectra of the 18 nm QW. Typical peaks of the charge-density, spin-density, and single-particle excitations were all observed with the proper polarization selection rule, labeled CDW, SDW, and SPE, respectively, and their energies were 56.0, 45.1, and 48.0 meV. Infrared absorption spectra could not be measured because GaAs substrates have strong absorption bands below 68 meV and prevented light transmission.

From these systematic results, we have concluded that intersubband electronic Raman scattering is dominated by single-particle excitations in narrow QWs, while intersubband infrared absorption shows collective charge-density excitation. Such a stark contrast between infrared absorption and resonant Raman scattering in narrow QWs has not been predicted before. Proper assignments of intersubband excitation peaks based on this are crucial in characterizing subband structures or investigating many-body Coulomb interaction effects.

To discuss why infrared absorption and resonant Raman scattering show such selection, or extremely different sensitivity, to collective and single-particle excitations, we make the following remarks.

Infrared absorption by the $E_0 \rightarrow E_1$ direct transition is electric-dipole allowed. In this process, incident photon energy is resonant with the subband separation at any electron in-plane wave vector $\mathbf{k}$, and thus all electrons with various $|\mathbf{k}| \leq k_F$ (k_F is the Fermi wave vector) form a collective excitation via many-body Coulomb interactions. This excitation involves a depolarization effect because the driving ac electric field is in a direction parallel to the quantum confinement. That is, infrared absorption occurs solely for collective charge-density excitation.

Electronic Raman scattering by the $E_0 \rightarrow H_n \rightarrow E_1$ transition, on the other hand, is electric-dipole forbidden for any virtual hole state H_n, because either of the $E_0 \rightarrow H_n$ and $H_n \rightarrow E_1$ interband transitions is forbidden by the parity selection rule in symmetric QWs. Enhancement by interband resonance is thus crucial in obtaining Raman signals with detectable intensities. Recently, Das Sarma and Wang have developed a theory of resonant Raman scattering in the random-phase approximation, considering parabolic subbands of the opposite signs for electrons and virtual holes. They have shown that the relative intensity of single-particle excitation to collective charge-density excitation is substantially enhanced under on-resonance-excitation conditions, and that the $\mathbf{k}$ dependence of the resonance factor is the key for the calculations.[10] This leads us to a physical interpretation that, when the incident photon energy is resonant with the interband energy gap between the E_1 and H_n subbands at one wave vector k_1 inside k_F, single-particle excitations at $|\mathbf{k}|=k_1$ are more enhanced than collective excitations (formed by all electrons with various $|\mathbf{k}| \leq k_F$).

Along the line of this insight, there are two points to be considered for the Raman peaks of collective excitations in wide QWs. First, k_F is relatively small in wide QWs where only the E_0 subband is populated. Thus, interband resonance affects most $|\mathbf{k}| \leq k_F$ simultaneously and enhances collective excitations in Raman processes. Second, the parity selection rule is partly broken in wide QWs because an asymmetric electric field occurs due to internal carriers, remote impurities caused by modulation doping, and surface charges. This enables nonresonant Raman processes to contribute to collective excitations.

Since the peak positions of collective charge-density excitation and single-particle excitation are obtained for the 10 nm QW as well as for wider QWs, we can investigate many-body Coulomb interaction energies in all three QWs. The collective charge-density and spin-density excitation energies can be expressed as[11–14]

$$E_{\mathrm{CD}} \approx E_{10} + (\alpha - \beta)N_S, \tag{1}$$

$$E_{\mathrm{SD}} \approx E_{10} - \beta N_S \tag{2}$$

in the local-density functional theory;[15–17] the single-particle excitation energy is equal to subband separation E_{10} including static many-body corrections.[4] αN_S and βN_S are dynamical many-body corrections called the depolarization shift and the excitonic shift due to the direct and exchange-correlation intersubband Coulomb interactions, respectively, and given by[11]

$$\alpha = \frac{e^2}{\kappa \epsilon_0} \int_{-\infty}^{\infty} dz \left(\int_{-\infty}^{z} dz' \zeta_0(z') \zeta_1(z') \right)^2, \tag{3}$$

$$\beta = -\int_{-\infty}^{\infty} dz\, \zeta_0(z)^2 \zeta_1(z)^2 \frac{\partial V_{xc}[n(z)]}{\partial n(z)}. \quad (4)$$

Here, e is the elementary charge, ϵ_0 is the vacuum permittivity, $\zeta_n(z)$ is the envelope wave function for the nth subband electron motion in the confinement direction (z direction), $V_{xc}[n(z)]$ is the exchange-correlation potential with $n(z) = N_S\zeta_0(z)^2$, and $\kappa = \kappa_\infty(E_{CD}^2 - E_{LO}^2)/(E_{CD}^2 - E_{TO}^2)$ with κ_∞ being the high-frequency dielectric constant, E_{LO} being the LO phonon energy, and E_{TO} being the TO phonon energy for background material. In the present cases, the contribution from phonons is small (i.e., $\kappa \approx \kappa_\infty$) because E_{CD} is much more than E_{LO}.

When $r_s = [4\pi a_0^3 n(z)/3]^{-1/3}$ is less than ~ 1, where $a_0 = \kappa_0 a_B/m^*$, a_B is the Bohr radius, and κ_0 is the static dielectric constant, we have[16,17]

$$V_{xc}[n(z)] \approx -\left(\frac{9\pi}{4}\right)^{1/3} \frac{e^2}{4\pi^2 \kappa_0 \epsilon_0 a_0 r_s} \quad (5)$$

and thus

$$\beta \approx \frac{(3\pi^2)^{1/3} e^2 N_S^{-2/3}}{12\pi^2 \kappa_0 \epsilon_0} \int_{-\infty}^{\infty} dz\, \zeta_0(z)^{2/3} \zeta_1(z)^2. \quad (6)$$

Table I shows values of the dynamical many-body effects calculated from Eqs. (3) and (6) for the three samples. The following material constants were used in the calculations: $E_{LO}=36.3$ meV, $E_{TO}=33.3$ meV, $\kappa_\infty=10.9$, and $\kappa_0=12.9$. Also shown in the table are values of $(\alpha-\beta)N_S$ obtained experimentally from the energy differences between the peaks of collective charge-density excitation and single-particle excitation. We found good agreement between the experimental and calculated values for $(\alpha-\beta)N_S$. Note that such an evaluation of the dynamical many-body effects over a wide range of E_{10} requires experimental values for narrow QWs, which can only be derived from the combination of absorption and Raman measurements.

The β/α of 0.29 for the 10 nm QW is relatively large despite the high electron concentration, compared with that for the 13.5 nm QW and those for wider QWs.[18] This shows that the excitonic shifts are more important for narrower QWs with a dependence of $\beta/\alpha \sim (N_S d_{eff}^2)^{-2/3}$, where d_{eff} is the effective thickness of the two-dimensional electron gas,[1,11] related to the well width.

Finally, we comment on the results presented in Fig. 2. So far, both infrared absorption and resonant Raman scattering for the parallel polarizations have been interpreted in theories as processes of the identical collective charge-density excitation, but the correspondence between their peaks has not been confirmed in experiments. Figure 2 gives direct evidence for the basic interpretation. It should also be noted that the linewidth of collective charge-density excitation observed in resonant Raman scattering (3.6 meV) was narrower than that in infrared absorption (5.4 meV).[19] Further investigations, particularly into resonant Raman scattering, by both experiments and theories are necessary for a microscopic understanding of linewidths as well as peak intensities.

In summary, we have directly compared intersubband infrared absorption and electronic Raman scattering in GaAs/AlAs single QWs with 10–18 nm width. The systematic change in their spectral features with well width clearly shows that resonant Raman scattering is dominated by single-particle excitations in narrow ($\sim$10 nm) QWs, while infrared absorption occurs solely for collective charge-density excitation. Analysis of dynamical many-body Coulomb interaction energies based on this fact suggests that the excitonic shifts are more important for narrower QWs, as explained in the local-density functional theory.

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*Electronic address: unuma@iis.u-tokyo.ac.jp
†Present address: Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan.

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