Large Exchange Interactions in the Electron Gas of GaAs Quantum Wells

A. Pinczuk, S. Schmitt-Rink, G. Danan, J. P. Valladares, L. N. Pfeiffer, and K. W. West
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

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Inelastic-light-scattering measurements show that exchange Coulomb interactions in the two-dimensional electron gas of GaAs microstructures are more important than previously anticipated. Small-wave-vector spectra from modulation-doped quantum wells exhibit unexpected single-particle intersubband transitions in addition to collective spin-density and charge-density modes. From the measured spectral energies the direct and exchange intersubband Coulomb interactions are determined and found to be of comparable strengths.

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Free electrons in semiconductor microstructures reveal new behaviors that arise from fundamental electron-electron interactions and the reduced dimensionality.[1] Strong correlations in the electron gas, as in the fractional quantum Hall effect, are revealed in magnetotransport[2] and also in magneto-optics.[3,4] More generally, Coulomb interactions and reduced dimensionality have strong manifestations in the elementary excitation spectrum of the free electrons. The energies and character of these excitations are directly studied in optical experiments, such as infrared absorption[5] and inelastic light scattering.[6]

The light-scattering method is especially powerful because both spin-density and charge-density excitations can be measured.[6,7] At small wave vectors the energies of spin-density modes are shifted from single-particle transition energies by the exchange Coulomb interaction.[1,8] Charge-density modes have energy shifts due to direct as well as exchange terms.[1,6-9] However, since exchange interactions were expected to be small in GaAs,[8] spin-density excitations were previously interpreted as the energy spacings of the quantum-well states and referred to as single-particle excitations.[7,10-12] Similarly, the shift of charge-density excitations from single-particle transition energies was considered in terms of direct Coulomb interactions and coupling to polar optical phonons.[6,7,10]

This Letter presents new results showing that such widely used interpretations of inelastic-light-scattering experiments require important revisions. In spectra of small-wave-vector intersubband excitations we find unexpected single-particle transitions in addition to the peaks of collective spin-density and charge-density modes. The significant shifts of the spin-density excitations from intersubband transition energies reveal large exchange interactions. With increasing wave vector, the collective spin-density and charge-density modes display enhanced broadening when their energies overlap the continua of single-particle transition (Landau damping). Analysis of the energies of the three excitations yields quantitative determinations of Coulomb interactions in which the exchange and direct terms have comparable strength. These observations highlight the role of electron-electron interactions in the physics of the electron gas in semiconductor microstructures.

The samples are GaAs-($Al_{0.3}Ga_{0.7}$)As quantum-well heterostructures grown by molecular-beam epitaxy. The well widths are 250 Å. Si dopants are introduced in the top barrier layer. The free electrons have extremely high mobilities in the range $(1-5)\times10^6$ $cm^2/V\,sec$ (at 2K) and their areal densities are within $(1.5-3)\times10^{11}$ cm^{-2}. Light-scattering spectra are measured in a conventional backscattering geometry.[6] The incident photon energies are resonant with excitonic optical transitions of the GaAs quantum wells.[13] Spectra are obtained with incident power densities below 1 W/cm^2 and as low as 10^{-2} W/cm^2. Optical multichannel detection is used in the acquisition of lowest intensity spectra. Spin-density excitations are active in depolarized spectra, when incident and scattered polarizations are orthogonal. Charge-density excitations occur in polarized spectra with parallel polarizations.

Figure 1 shows spectra of intersubband excitations obtained with high resolution (0.06 meV$\simeq$15 GHz). The illuminated area of the sample is a circle of radius about 50 μm. The sharp peaks that have well-defined polarization selection rules are assigned to collective spin-density excitations (SDE) and charge-density excitations (CDE). They are shifted from the spacing between the two lowest-conduction subbands by the effects of direct and exchange Coulomb interactions. These peaks are the sharpest intersubband excitations ever reported in GaAs. The line shapes are not changed when the illuminated area is increased by a factor of 10. This indicates that inhomogeneous broadening is not important in these measurements. These results are typical of the high-electron-mobility quantum wells studied.

The spectra of Fig. 1 also show unexpected bands labeled SPE (single-particle excitations). To identify the elementary excitations associated with these features we studied the dependence on the in-plane scattering wave vector k. Results from a higher-density sample are

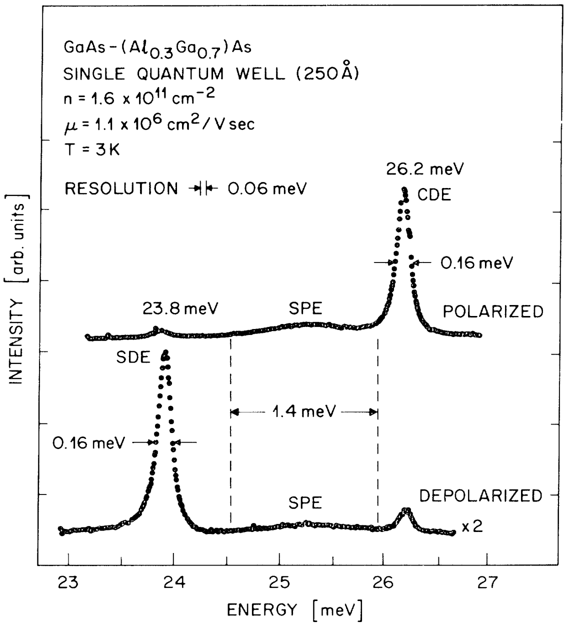

FIG. 1. Inelastic-light-scattering spectra of intersubband excitations of the high-mobility 2D electron gas in a GaAs quantum well. The peaks of spin-density excitations (SDE), charge-density excitations (CDE), and single-particle excitations (SPE) are shown.

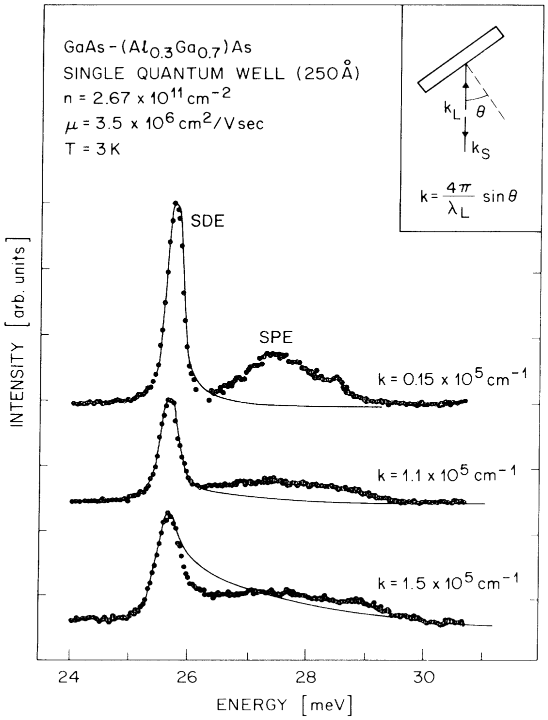

FIG. 2. Depolarized light-scattering spectra of intersubband excitations for several values of the scattering wave vector k. The lines are fits of SDE spectra using Eqs. (1)-(4). Inset: The scattering geometry and the expression for k.

presented in Figs. 2 and 3. Here, the spectral resolution is 0.15 meV. The inset to Fig. 2 shows the geometry of the experiment. The depolarized spectra in Fig. 2 reveal that the width of the SPE band has a marked dependence on in-plane scattering wave vector. Such k dependence is expected for nonvertical single-particle intersubband transitions. In the long-wavelength limit $k \ll k_F$, the energies of these transitions, as sketched in the inset to Fig. 3, cover a continuum bounded by $E_{01} \pm k v_F$, where v_F is the Fermi velocity and E_{01} is the subband spacing at $k=0$. At the large wave vectors, the width of the SPE bands is indeed about $2kv_F$. The k dependence identifies the SPE features as single-particle intersubband transitions centered at E_{01}.

This assignment explains the changes in the line shape of the charge-density collective mode in the spectra of Fig. 3. With increasing k CDE overlaps the continuum of single-particle excitations, as shown in the inset to Fig. 3. This causes the decay of CDE into intersubband electron-hole pairs (Landau damping). The linewidths of the collective spin-density modes in the spectra of Fig. 2 also increase with k. The effect is less pronounced in this case because the $k \sim 0$ SDE is further away from

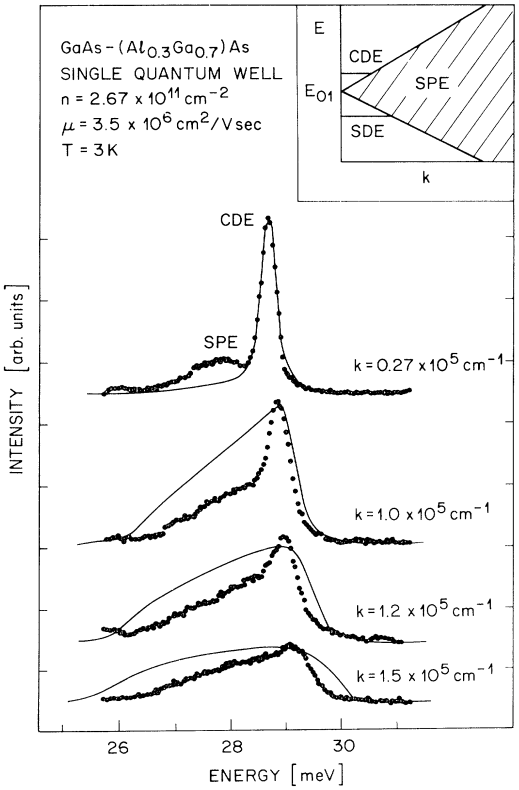

FIG. 3. Polarized light-scattering spectra of intersubband excitations for several values of the scattering wave vector k. The lines are fits of CDE spectra using Eqs. (1)-(4). Inset: A sketch of the k dependence of intersubband excitations in the long-wavelength limit $k \ll k_F$.

E_{01}. This result shows that spin-density excitations are subject to Landau damping and demonstrates their collective character.

The assignment of the SPE to single-particle intersubband transitions allows measurements of E_{01}. The inset to Fig. 3 shows that for finite k the single-particle continuum is (i) centered at E_{01} and (ii) symmetric about this energy. The spectra of Fig. 2, where Landau damping has only a minor effect on the line shape, show that the SPE bands broaden indeed symmetrically with increasing k. This identifies E_{01} as the peak position of the SPE band. The uncertainties in these determinations of E_{01} are less than 0.25 meV.

The measurement of E_{01} in addition to that of the energies of SDE and CDE, ω_{SD} and ω_{CD}, allows direct determinations of Coulomb interactions in intersubband transitions. In the analysis we write the spectral intensities as

$$I_j(k,\omega) \sim \operatorname{Im}\chi_j(k,\omega)\,. \qquad (1)$$

In the response functions for intersubband excitations, $\chi_j(k,\omega)$, we use phenomenological k-dependent local-field corrections similar to those obtained within generalized random-phase approximations (RPA),[8,9]

$$\chi_j(k,\omega) = \frac{\chi_0(k,\omega)}{1-\gamma_j(k)\chi_0(k,\omega)}\,, \qquad (2)$$

where $\chi_0(k,\omega)$ is the intersubband susceptibility.[8,9,14-16] For $k=0$, Eqs. (1) and (2) yield peaks at energies $\omega_j^2 = E_{01}^2 + 2n\gamma_j(0)E_{01}$. For spin-density excitations we set $\gamma_{\mathrm{SD}}(0) = -\beta_{01}$, where β_{01} is a positive parameter that accounts for the exchange Coulomb interaction, i.e., the "excitonic" effect between the electron in the excited subband and the hole in the ground state. For charge-density excitations $\gamma_{\mathrm{CD}}(0) = \alpha_{01}/\epsilon(\omega) - \beta_{01}$, where α_{01} is the direct ("depolarization") term and $\epsilon(\omega)$ is the dielectric function of the polar lattice. For α_{01} and β_{01} we obtain

$$\frac{E_{01}^2-\omega_{\mathrm{SD}}^2}{E_{01}} = 2n\beta_{01}\,, \qquad (3)$$

$$\frac{\omega_{\mathrm{CD}}^2-\omega_{\mathrm{SD}}^2}{E_{01}}\epsilon(\omega_{\mathrm{CD}}) = 2n\alpha_{01}\,. \qquad (4)$$

In the present study we consider α_{01} and β_{01} as parameters that are obtained from the measured energies of intersubband excitations using Eqs. (3) and (4). Figure 4 shows the results from four quantum wells of width 250 Å. Also shown are the results from a higher-density sample of well width 200 Å. While the values of $2n\alpha_{01}$ are comparable to those previously reported,[10] those of $2n\beta_{01}$ indicate exchange interactions considerably larger than anticipated. Clearly, the two Coulomb interactions have comparable strength. For $n \lesssim 3\times10^{11}$ cm^{-2} their ratio is approximately independent of electron density and equals $\beta_{01}/\alpha_{01} \simeq 0.4$. An estimate based on local-spin-density-functional theory predicts a smaller ratio of $\beta_{01}/\alpha_{01} \simeq 0.2$.[8]

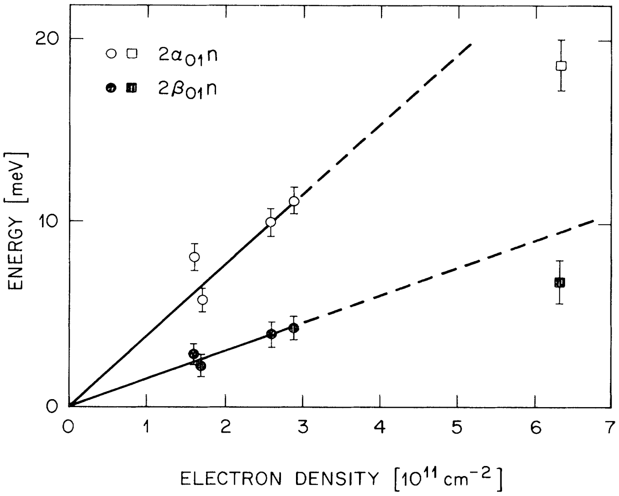

FIG. 4. Density dependence $2\alpha_{01}n$ and $2\beta_{01}n$ obtained from measured spectral energies using Eqs. (3) and (4). The dots are from 250-Å wells and the squares from a 200-Å well. Lines are a guide to the eye.

Figures 2 and 3 also show one-parameter theoretical fits to the experimental spectra, based on Eq. (2) convoluted with a Lorentzian of k-independent width. Although the experimental k's are smaller than the Fermi wave vector (but comparable to the inverse well width), the $\gamma_j(k)$'s are found to decrease with k. This should be contrasted with the k independent β_{01} resulting from local-spin-density-functional theory.[8] A constant β_{01} would yield a stronger dispersion of the SDE than observed experimentally. The fits reproduce the overall behavior of the collective excitations. The deviations at larger k (the experimental SDE and CDE are somewhat sharper than the theoretical ones) could be attributed to several experimental factors like microscopic (<10 μm) variations in electron density and the geometry of the experiment which tends to emphasize small-angle scattering. On the theoretical side, a frequency-independent local-field correction might not accurately model the solution of the Bethe-Salpeter equation for the particle-hole propagator away from $k=0$.

The one-parameter fits do not account for the unexpected SPE structures in the small-k data. It is presently unclear to us which light-scattering processes are at work. Fermi sea "shake-up," in which intersubband and intrasubband excitations would be simultaneously created, does not appear to produce such spectra.[17,18] This leaves us with elastic scattering due to residual disorder as the most likely candidate. In fact, in high magnetic fields, disorder-induced breakdown of wave-vector conservation has been clearly observed in light scattering from high-mobility GaAs quantum wells.[19] Similar processes could accommodate the wave vectors $\sim5\times10^4$ cm^{-1} required to explain the widths of the SPE bands in zero field.

In conclusion, light-scattering spectra were used to determine for the first time the absolute magnitude of exchange intersubband Coulomb interactions in the electron gas of GaAs quantum wells. Contrary to widespread belief, exchange and direct terms are found to be of comparable strength. The ground-state intrasubband exchange interaction is expected to be of a similar magnitude, which casts some doubt on RPA-based theories of the two-dimensional electron gas. We also note that an exchange enhancement of the size reported here was postulated earlier to explain certain polarization anomalies in quantum-well luminescence.[20]

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[1] T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).

[2] D. C. Tsui and H. L. Störmer, IEEE J. Quantum Electron. **22**, 1711 (1986).

[3] I. V. Kukushkin and V. B. Timofeev, Pis'ma Zh. Eksp. Teor. Fiz. **44**, 179 (1986) [JETP Lett. **44**, 228 (1986)].

[4] D. Heiman, B. B. Goldberg, A. Pinczuk, C. W. Tu, A. C. Gossard, and J. H. English, Phys. Rev. Lett. **61**, 605 (1988).

[5] W. Hansen, M. Horst, J. P. Kotthaus, U. Merkt, Ch. Sikorski, and K. Ploog, Phys. Rev. Lett. **58**, 2586 (1987).

[6] G. Abstreiter, R. Merlin, and A. Pinczuk, IEEE J. Quantum Electron. **22**, 1771 (1986).

[7] E. Burstein, A. Pinczuk, and D. L. Mills, Surf. Sci. **98**, 451 (1980).

[8] T. Ando, J. Phys. Soc. Jpn. **51**, 3893 (1982); S. Katayama and T. Ando, *ibid.* **54**, 1615 (1985).

[9] A. C. Tselis and J. J. Quinn, Phys. Rev. B **29**, 3318 (1984); G. Eliasson, P. Hawrylak, and J. J. Quinn, *ibid.* **35**, 5569 (1987).

[10] A. Pinczuk and J. M. Worlock, Surf. Sci. **113**, 69 (1982).

[11] D. Y. Oberli, D. R. Wake, M. V. Klein, J. Klem, T. Henderson, and H. Morkoc, Phys. Rev. Lett. **59**, 696 (1987).

[12] K. Bajema, R. Merlin, F. Y. Yuang, S. C. Hong, J. Singh, and P. K. Bhattacharya, Phys. Rev. B **36**, 1300 (1987).

[13] G. Danan, A. Pinczuk, J. P. Valladares, L. N. Pfeiffer, K. W. West, and C. W. Tu, Phys. Rev. B **39**, 5512 (1989).

[14] L. Wendler and R. Pechstedt, Phys. Rev. B **35**, 5887 (1987).

[15] D. H. Ehlers, Phys. Rev. B **38**, 9706 (1988).

[16] D. A. Dahl and L. J. Sham, Phys. Rev. B **16**, 651 (1977).

[17] C. S. Ting and A. K. Ganguly, Phys. Rev. B **20**, 4244 (1979).

[18] There is a close relationship between the present work and the still unsolved problem of interband absorption in doped semiconductors. See, e.g., J. Gavoret, P. Nozieres, B. Roulet, and M. Combescot, J. Phys. (Paris) **30**, 987 (1969); A. E. Ruckenstein and S. Schmitt-Rink, Phys. Rev. B **35**, 7551 (1987).

[19] A. Pinczuk, J. P. Valladares, D. Heiman, A. C. Gossard, J. H. English, C. W. Tu, L. N. Pfeiffer, and K. W. West, Phys. Rev. Lett. **61**, 2701 (1988).

[20] A. E. Ruckenstein, S. Schmitt-Rink, and R. C. Miller, Phys. Rev. Lett. **56**, 504 (1986).