Absence of unstable zero-field intersubband spin excitations of dilute electron bilayers

A. S. Plaut
Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974
and Department of Physics, University of Exeter, Exeter EX4 4QL, United Kingdom

A. Pinczuk
Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974

P. I. Tamborenea
Institut für Theoretische Physik, Universität Frankfurt, D-60054 Frankfurt am Main, Federal Republic of Germany

B. S. Dennis, L. N. Pfeiffer, and K. W. West
Bell Laboratories, Lucent Technologies, Murray Hill, New Jersey 07974
(Received 26 December 1996)

Low-energy spin-density intersubband excitations of electron bilayers in GaAs double quantum wells have been investigated by inelastic light scattering in the search for unstable modes. Excitonic vertex corrections due to exchange Coulomb interactions were thus determined down to very low electron densities. The experiments are interpreted within the time-dependent local-density approximation (TDLDA) and with a nonlocal theory. The pronounced softening at zero magnetic field (a vertex-correction driven instability) predicted by the TDLDA for densities $n<10^{11}$ cm^{-2} was not observed. At these lower densities we achieve better agreement with experiment using the nonlocal theory. [S0163-1829(97)04215-X]

Two-dimensional (2D) electron gases formed at GaAs/$Al_xGa_{1-x}As$ heterojunctions are particularly appropriate for studies of unusual electron behavior, as impurity scattering is minimized due to the spatial separation of the mobile electrons from their donors. Owing to rapid progress in the epitaxial growth of heterojunctions, it is now possible to carry out a diverse group of experiments that probe the fundamental electron interactions in low-dimensional systems.[1] The striking physics arising from the Coulomb interaction is manifested in the energies and dispersions of collective modes constructed from neutral particle-hole pairs in the 2D electron systems. In the regimes of the integer and fractional quantum Hall effects excitonic bindings from vertex corrections due to the exchange terms of the Coulomb interaction are responsible for characteristic magnetoroton minima in the calculated dispersions of collective excitations.[2] Similar magnetoroton minima are predicted for charge-density intersubband excitations (CDEs) across the tunneling gap of electron bilayers in strong perpendicular magnetic fields.[3] The mode softenings associated with such vertex-correction driven magnetoroton minima are the source of instabilities that explain phase transitions in coupled double quantum wells (DQWs) at Landau level filling $\nu=1$.[3,4]

Very recently, a different class of phase transition has been predicted to occur in electron bilayers in DQWs in the absence of magnetic fields.[5] Here the instability is caused by a vertex-correction driven softening of the lowest $\mathbf{q}=0$ spin-density intersubband excitation (SDE) of the lowest symmetric and antisymmetric states. This is readily seen in the expression for the mode energy[6]

$$\omega_{\mathrm{SDE}}^2=\Delta_{\mathrm{SAS}}^2-2(n_S-n_{\mathrm{AS}})\beta\Delta_{\mathrm{SAS}}, \qquad (1)$$

where n_S and n_{AS} are the populations of the symmetric and antisymmetric subbands, respectively, and Δ_{SAS} is the symmetric-antisymmetric tunneling gap shown in the inset to Fig. 1. The factor (n_S-n_{AS}) takes into account the reduction in available phase space when two subbands are occupied. The second term on the right-hand side of Eq. (1) represents the excitonic binding and the factor β parametrizes the strength of the vertex corrections. When this intrawell excitonic term is larger than the interwell couplings represented by Δ_{SAS} the tunneling gap collapses for the intersubband SDE. The possible existence of such unstable spin-density excitations suggests the emergence of broken-symmetry phases of antiferromagnetically correlated 2D layers. The time-dependent local-density approximation (TDLDA) results of Ref. 5 predict phase transitions for electron densities below 10^{11} cm^{-2}.

In this paper we present the results of a comprehensive search for the soft $\mathbf{q}=0$ intersubband SDEs that could lead to a broken-symmetry phase in DQWs at zero magnetic field. Resonant inelastic light-scattering measurements of the SDE modes have been carried out on a large set of GaAs DQWs of various well shapes and with electron densities down to 10^{10} cm^{-2}. The sample parameters were chosen[7] to overlap those for which TDLDA calculations indicate the existence of an unstable SDE that triggers transitions to broken-symmetry phases.

These experiments, however, reveal only a weak dependence of low-energy SDE modes on electron density, in striking disagreement with the predictions of TDLDA theory. The results are intriguing because pronounced softenings of low-energy $\mathbf{q}=0$ SDEs, in general agreement with Eq. (1), are seen in spectra from DQWs in perpendicular magnetic fields at even values of ν.[8] The discrepancies be-

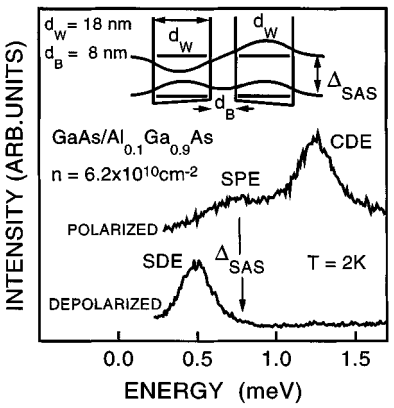

FIG. 1. Inelastic light-scattering spectra of the intersubband excitations of the double quantum well. The peaks of the spin-density excitations (SDE), charge-density excitations (CDE), and single-particle excitations (SPE) are shown. The spectra have been offset for clarity. Inset: schematic of the symmetric and antisymmetric states of the DQW structures.

tween experiment and theory are attributed here to shortcomings of the TDLDA calculation at low densities, when local-density approximations become unreliable because very dilute electron gases can no longer be considered homogeneous even on a local scale. This conclusion is supported by a calculation that treats the exchange Coulomb interaction by means of a variational solution of the Bethe-Salpeter equation in the ladder approximation.[9] Such a nonlocal approach yields predictions for the energies of intersubband collective modes in the DQW that are in good agreement with experiment.

The modulation-doped DQWs were grown by molecular-beam epitaxy. They consist of two nominally identical GaAs quantum wells of width $d_w = 180$ Å separated by either 40-Å $Al_{0.2}Ga_{0.8}As$ barriers or 80-Å $Al_{0.1}Ga_{0.9}As$ barriers (inset to Fig. 1). Si δ-doped layers grown both above and below the DQW create two equivalent 2D electron gases with combined total carrier concentrations in the range 0.15×10^{11} cm^{-2}<n<1.7×10^{11} cm^{-2} (determined from both transport[7] and optical measurements[10]) and low-temperature mobilities $(0.2-1.5)\times10^{6}$ cm^2/V s. Resonant inelastic light-scattering spectra were obtained in a conventional backscattering geometry at 1.7 K using a dye laser tuned to the optical transitions of the GaAs quantum wells.

Figure 1 shows the lowest-energy intersubband spectra obtained from a sample with an $Al_{0.1}Ga_{0.9}As$ barrier. For $\mathbf{q} \simeq 0$, the energy of the single-particle excitations (SPEs) equals that of Δ_{SAS}. The highest energy peak arises from the CDEs. As the depolarization shift due to the direct terms of the Coulomb interactions is usually greater than the excitonic shift, the CDEs appear higher in energy than both the SPE's and SDEs.[6] The collective SDEs and CDEs display well-defined polarization selection rules. SDEs are active in depolarized spectra, where incident and scattered light polarizations are perpendicular and CDEs occur in polarized spectra measured with parallel polarizations. The sharpness of the collective excitation peaks is indicative of the high quality of the bilayers.

We have measured the energy of the CDE, SDE, and SPE modes in both series of samples as a function of carrier concentration and obtained similar results from both series. In this paper we will concentrate on the results for the samples with $Al_{0.1}Ga_{0.9}As$ barriers, which we have plotted in Fig. 2(a). To interpret these experiments we have calculated the collective intersubband excitation energies using two different approaches: The first is the TDLDA (Refs. 5 and 11) and the second is a variational solution of the Bethe-Salpeter equation in the ladder approximation.[9] While the TDLDA treats the exchange-correlation interaction in a local fashion, assuming a point-contact Coulomb interaction, the latter, nonlocal, theory takes the exchange interaction into account exactly.

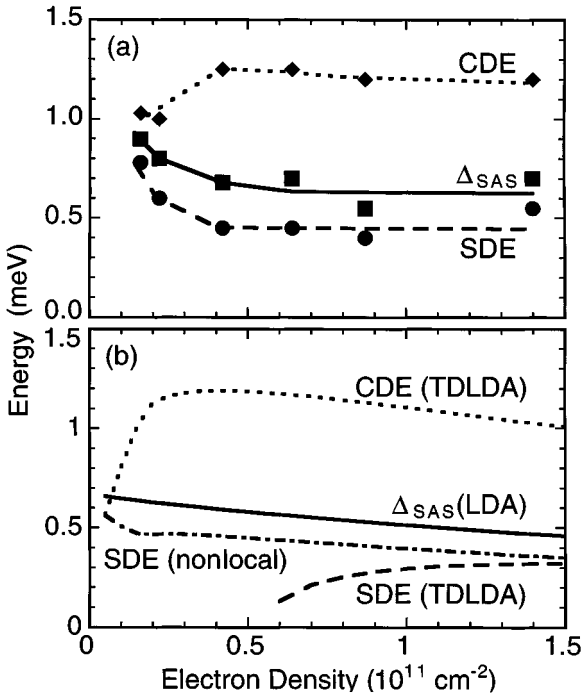

FIG. 2. Intersubband charge-density excitation energy ω_{CDE} (dotted line), spin-density excitation energy ω_{SDE} (dashed and dot-dashed lines), and single-particle excitation energy Δ_{SAS} (solid line) as a function of the two-dimensional electron density for the GaAs/$Al_{0.1}Ga_{0.9}As$ double-quantum-well structures. (a) Measured values (the lines are just guides to the eye) and (b) values calculated self-consistently within a time-dependent local-density approximation (TDLDA) and using a nonlocal approach (dot-dashed line).

In the TDLDA, the vertex correction β is given by

$$\beta_{\mathrm{LDA}} = -\int_{-\infty}^{\infty} dz\ \varphi_1(z)\varphi_0(z)\frac{\partial V_{\mathrm{xc}}}{\partial n}(z)\varphi_1(z)\varphi_0(z), \quad (2)$$

where $\varphi_i(z)$ are the LDA-calculated self-consistent wave functions and V_{xc} is the exchange-correlation potential.[12] In the nonlocal theory, one obtains

$$\beta_{\mathrm{NL}} = \frac{2}{(n_S - n_{\mathrm{AS}})^2}\int \frac{d^2k_1 d^2k_2}{(4\pi)^4}\,\theta(k_{F,0}-k_1)\,\theta(k_1-k_{F,1}) \times V^{s}_{1100}(\mathbf{k}_1-\mathbf{k}_2)\,\theta(k_{F,0}-k_2)\,\theta(k_2-k_{F,1}), \quad (3)$$

where V^{s}_{1100} is an intersubband matrix element of the screened Coulomb interaction, calculated within the static random-phase approximation. Equation (3) is valid with either one or two subbands occupied; in the former case, one

must set the Fermi wave vector of the second subband $k_{F,1}=0$. The CDE energy is given by

$$\omega_{\mathrm{CDE}}^2=\omega_{\mathrm{SDE}}^2-2(n_S-n_{\mathrm{AS}})\alpha\Delta_{\mathrm{SAS}}, \tag{4}$$

where α is the depolarization shift, given by

$$\alpha=-\frac{4\pi e^2}{\epsilon}\int_{-\infty}^{\infty}dz\ \varphi_1(z)\varphi_0(z) \times\int_{-\infty}^{z}dz'\int_{-\infty}^{z'}dz''\varphi_1(z'')\varphi_0(z''). \tag{5}$$

Here $\epsilon\simeq\epsilon(0)$ is the low-frequency dielectric constant. Figure 2(b) shows the results of both theoretical approaches.

A striking feature of the theoretical evaluations, displayed in Fig. 2(b), is the vanishing SDE mode energy predicted by the TDLDA calulation for electron densities below 0.5×10^{11} cm^{-2}. Such behavior, caused by an enhanced vertex-correction induced excitonic shift, signals an instability in the Fermi liquid that triggers a phase transition to a many-body excitonic liquid ground state.[5] This prediction of the LDA is absent in the experimental results of Fig. 2(a). The results of the nonlocal calculation in Fig. 2(b), on the other hand, are in good agreement with the measured SDE mode energies [Fig. 2(a)].

Other features predicted by the LDA calculation are observed experimentally. Figure 2 shows good agreement between the experimental and the LDA values of SPE and CDE energies. The experimental SPE energies show the upward trend with decreasing electron density expected from the renormalized Δ_{SAS} calculated self-consistently within LDA.[13] In addition, the calculation predicts that at $n<10^{10}$ cm^{-2} the excitonic vertex correction should become larger than the depolarization shift resulting in the energy of the CDEs falling below that of the SPEs.[11,14] Indeed, in the experimental data a similar decrease in the CDE energy is observed as n approaches 10^{10} cm^{-2}. As will be seen below, this is due to a decrease in the depolarization shift rather than an increase in the excitonic shift, which is again in qualitative agreement with LDA theory.[11]

To obtain further insight into the many-body interactions we rewrite Eqs. (1) and (4) to determine the direct and exchange terms of the Coulomb interaction

$$(n_S-n_{\mathrm{AS}})\alpha=\frac{\omega_{\mathrm{CDE}}^2-\omega_{\mathrm{SDE}}^2}{2\Delta_{\mathrm{SAS}}}, \tag{6}$$

$$(n_S-n_{\mathrm{AS}})\beta=\frac{\Delta_{\mathrm{SAS}}-\omega_{\mathrm{SDE}}^2}{2\Delta_{\mathrm{SAS}}}. \tag{7}$$

The depolarization shift $(n_S-n_{\mathrm{AS}})\alpha$ and the excitonic shift $(n_S-n_{\mathrm{AS}})\beta$ are derived from the measured mode energies shown in Fig. 2(a). These results are plotted along with the theoretical evaluations in Fig. 3. In Fig. 3(b) we find that the LDA predicts that $(n_S-n_{\mathrm{AS}})\alpha$ increases slightly with decreasing n and then that it drops sharply for electron densities below 0.2×10^{11} cm^{-2}. The maximum corresponds to the density at which the second subband just becomes occupied. We do indeed observe this behavior experimentally, as is shown in Fig. 3(a). Thus the LDA seems to work well when describing the depolarization shift down to very low electron densities.

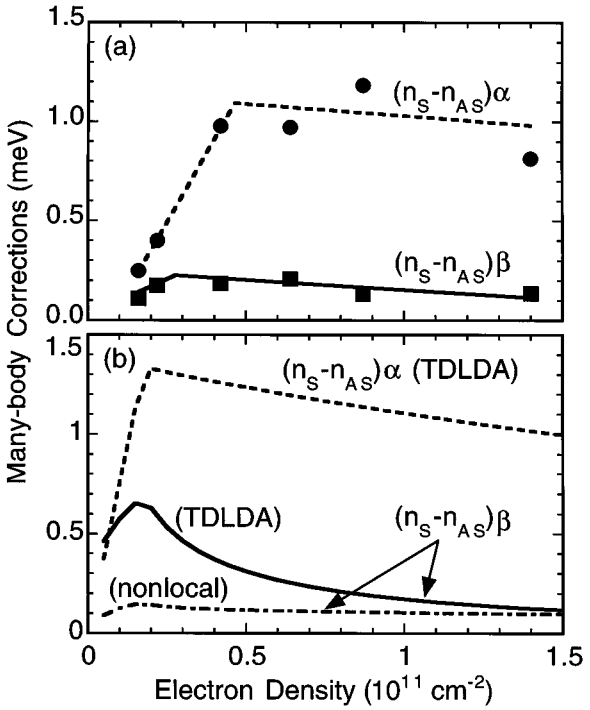

FIG. 3. Density dependence of $(n_S-n_{\mathrm{AS}})\alpha$ (dashed line) and $(n_S-n_{\mathrm{AS}})\beta$ (solid and dot-dashed lines) for the GaAs/$Al_{0.1}Ga_{0.9}As$ double-quantum-well structures, (a) obtained from measured spectra (the lines are just guides to the eye) and (b) calculated self-consistently within a TDLDA and using a nonlocal theory (dot-dashed line).

A different situation prevails in the case of the vertex correction $(n_S-n_{\mathrm{AS}})\beta$ because here we find significant quantitative differences between experiment and LDA theory. In fact, as seen in Fig. 3, for electron densities below 0.5×10^{11} cm^{-2} the experimental values of $(n_S-n_{AS})\beta$ are 2-5 times smaller than the predictions of the TDLDA calculation. The most significant discrepancies occur at densities below 0.2×10^{11} cm^{-2}, close to the onset of occupation of the antisymmetric state. Thus the TDLDA calculation is not successful at describing the observed behavior of both the vertex corrections and the SDE mode energies at very low density. On the other hand, the results of Fig. 3 show that the nonlocal calculation gives remarkably good agreement with our experimental findings.

The kind of disagreement between TDLDA and experiment that we see here has probably not been seen before because previous studies[9] considered single quantum wells, which have, in general, less inhomogeneous electron densities than DQWs. In a DQW structure the electronic density has rapid variations, due to the presence of the extra middle barrier, which may render the local-density approximation inapplicable since it requires small density gradients. The nonlocal solution of the Bethe-Salpeter equation is exact in this sense and takes into account the exact Coulomb exchange interaction of the inhomogenous system. Also, in single wells, the subband separations are much larger than in this study and therefore the small difference between

TDLDA and nonlocal treatments was relatively unimportant.[9] In addition, the TDLDA calculation relies on an estimate of the exchange interaction based on numerical evaluations of the ground-state energy of a three-dimensional electron gas. Probably, the exchange energy calculated for a uniform three-dimensional electron gas overestimates the exchange interaction of a two-component electron system since the additional quantum number (subband number) reduces the influence of the Pauli exclusion principle. Therefore, the vertex correction is overestimated in the TDLDA and the Bethe-Salpeter equation gives a more accurate result.

Before concluding, we comment on the marked softenings of the $\mathbf{q}=0$ intersubband SDEs observed in dilute electron bilayers of DQWs at even integer values of ν.[8] The striking enhancement of the vertex corrections in magnetic field suggests possible mechanisms for the emergence of broken-symmetry phases driven by unstable $\mathbf{q}=0$ spin-density modes.[8,15] The absence of zero-field enhancements of the vertex correction $(n_S-n_{\mathrm{AS}})\beta$, determined above, indicates that the softenings of the spin-density mode in perpendicular field are due to changes in the effective density of electrons that contribute to the intersubband collective excitations. At zero field and at small values of Δ_{SAS}, the effective electron density, given by (n_S-n_{AS}), is limited by the occupation of the antisymmetric state. However, this effective density can be changed by the magnetic field, and for filling factor $\nu=2$ and lower all the electrons may contribute.

Thus, in conclusion, our detailed light-scattering study of low-energy spin-density excitations of modulation-doped $\mathrm{GaAs/Al}_x\mathrm{Ga}_{1-x}\mathrm{As}_x$ DQWs shows the absence of soft modes that could trigger a phase transition in the electron system at zero magnetic field. In these experiments we determined the single-particle tunneling gaps Δ_{SAS}, and the corrections to intersubband collective energies due to Coulomb interactions. The measurements are compared to those calculated using the TDLDA and a nonlocal (Bethe-Salpeter) theory. We find that in dilute electron double layers the nonlocal theory offers a good description of the measured excitonic shifts due to exchange interactions, while the local-density approach overestimates these vertex corrections.

We acknowledge many substantial contributions of J. P. Eisenstein in the early stages of this work. We thank the authors of Ref. 15 for discussions and for letting us know of their results prior to publication. A.S.P. gratefully acknowledges financial support from The Leverhulme Trust, United Kingdom.

[1] Proceedings of the 11th International Conference on the Electronic Properties of Two Dimensional Systems, 1996 [Surf. Sci. **361/362**, and references therein].

[2] C. Kallin and B. I. Halperin, Phys. Rev. B **30**, 5655 (1984); F. D. M. Haldane and E. H. Rezayi, Phys. Rev. Lett. **54**, 237 (1985); S. Girvin, A. H. MacDonald, and P. M. Platzman, *ibid.* **54**, 581 (1985).

[3] H. A. Fertig, Phys. Rev. B **40**, 1087 (1989); A. H. MacDonald, P. M. Platzman, and G. S. Boebinger, Phys. Rev. Lett. **65**, 775 (1990); L. Brey, *ibid.* **65**, 903 (1990).

[4] X. M. Chen and J. J. Quinn, Phys. Rev. B **45**, 11 054 (1992); R. Cote, L. Brey, and A. H. MacDonald, *ibid.* **46**, 10 239 (1992); Song He, S. Das Sarma, and X. C. Xie, *ibid.* **47**, 4394 (1993).

[5] S. Das Sarma and P. I. Tamborenea, Phys. Rev. Lett. **73**, 1971 (1994); R. J. Radtke and S. Das Sarma, Solid State Commun. **96**, 215 (1995); **98**, 771 (1996).

[6] R. Decca *et al.*, Phys. Rev. Lett. **72**, 1506 (1994).

[7] G. S. Boebinger *et al.*, Phys. Rev. Lett. **64**, 1793 (1990); J. P. Eisenstein *et al.*, *ibid.* **68**, 1383 (1992); J. P. Eisenstein, L. N. Pfeiffer, and K. W. West, *ibid.* **69**, 3804 (1992); S. Q. Murphy *et al.*, *ibid.* **72**, 728 (1994).

[8] A. Pinczuk *et al.*, Bull. Am. Phys. Soc. **41**, 482 (1996); A. Plaut, *ibid.* **41**, 590 (1996); V. Pellegrini *et al.*, Phys. Rev. Lett. **78**, 310 (1997).

[9] J. C. Ryan, Phys. Rev. B **43**, 12 406 (1991); D. Gammon *et al.*, Phys. Rev. Lett. **68**, 1884 (1992); S. L. Chuang *et al.*, Phys. Rev. B **46**, 1897 (1992).

[10] A. Pinczuk *et al.*, Solid State Commun. **50**, 735 (1984).

[11] S. Das Sarma and I. K. Marmorkos, Phys. Rev. B **47**, 16 343 (1993); I. K. Marmorkos and S. Das Sarma, *ibid.* **48**, 1544 (1993).

[12] D. M. Ceperley and B. J. Alder, Phys. Rev. Lett. **45**, 566 (1980); J. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981); T. Ando, A.B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982). In the calculation of the vertex correction of Eq. (2) for spin-density excitations, dV_{xc}/dn is replaced by dV_{xc}/dm, where m is the spin density. The numerical differences between these two expressions are minor in our problem, and we have used dV_{xc}/dn for both types of excitations.

[13] A. S. Plaut *et al.*, Solid State Electron. **40**, 291 (1996).

[14] S. Ernst *et al.*, Phys. Rev. Lett. **72**, 4029 (1994).

[15] L. Zheng, R. J. Radtke, and S. Das Sarma (unpublished).