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Nonlinear dependence of the magnetophotoluminescence energies of asymmetric GaAs/$Ga_{0.67}Al_{0.33}As$ quantum wells on an external magnetic field

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Nonlinear dependence of the magnetophotoluminescence (MPL) energies in modulation-doped asymmetric GaAs/$Ga_{0.67}Al_{0.33}As$ quantum wells of different widths are investigated experimentally and theoretically as functions of an external magnetic field. The investigated structures have only one electric subband populated with electrons. Contrary to the theoretical descriptions existing in the literature and based on the oscillations of screening, the observed maxima of MPL energies do not occur at integer filling factors and do not change into minima for higher well widths. We interpret our observations assuming that the oscillations of MPL energies are due to an oscillatory electron transfer between a GaAs well and a reservoir outside the well. We obtain a very good description of the experimental data concerning both the maxima positions and the oscillation amplitudes for different well widths and electron densities. Our interpretation is corroborated by the quantum Hall data obtained on the same samples.

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I. INTRODUCTION

From the early days of optical experiments with semiconductor heterostructures, it was observed that the energies of interband magnetophotoluminescent (MPL) transitions exhibit striking nonlinear behavior as functions of an external magnetic field.[1] Such nonlinearities are characteristic of two-dimensional (2D) systems and are not seen on bulk materials. Since the first observations, the nonlinear behavior became the subject of numerous experimental and theoretical investigations, persisting until today.

The investigated systems can be divided into two categories. The first consists of structures in which, for reasons of material composition, electron density, or the shape of the quantum well (QW), more than one conduction subband is populated with electrons.[2–9] The nonlinear MPL energies observed on such structures were consistently explained by an electron transfer between the subbands in question. As an external magnetic field is increased, the oscillating Landau density of states at the Fermi energy is shifted which may cause the electron transfer between electric subbands. Since higher subbands have a larger spatial extension than lower subbands, the electron transfer changes the charge distribution in the well. This process affects the corresponding electric potential (the band bending) and, in turn, the subband energies. Thus, the whole system "breathes" periodically as the field is swept, which is reflected in the MPL energies.

The situation is different if one deals with a system in which only one subband is occupied. An important example of such a situation is a rather narrow and not strongly doped GaAs/$Ga_{0.67}Al_{0.33}As$ quantum well. In this case one cannot explain the nonlinearities by the above mechanism. Two theoretical calculations for this situation were proposed and both reached similar conclusions.[10,11] As the consecutive Landau levels (LL) cross the Fermi energy in an increasing magnetic field, the oscillatory density of states gives rise to oscillations of screening. These oscillations result in the oscillations of the gap renormalization which is repeated by the interband energies. For symmetric QWs, the MPL energies should show positive spikes at even filling factors. Tsuchiya *et al.*[12] extended this work to asymmetric QWs. Such wells allow one to separate in the real space electrons and holes which influences the screening of photogenerated holes by the 2D gas. The calculation predicted that, for a 10 nm wide QW, when the electrons and holes are spatially not well separated, one should observe positive spikes in the interband energies. On the other hand, for a 30 nm wide QW, in which electrons and holes are much better separated, the screening effects for holes are much smaller. As a result, the theory predicted for this situation negative spikes of interband energies at even filling factors. For the intermediate width of $d=20$ nm the theory predicted almost no spikes.

In a more recent work, Asano and Ando [13,14] investigated theoretically MPL energies in both symmetric and asymmetric QWs using a numerical diagonalization method. The results were obtained in the form of numerous δ functions which had to be phenomenologically broadened in order to represent MPL peaks. In Refs. 13 and 14, the authors could not confirm the phase reversal of peaks as a function of the well width predicted in Ref. 12.

It should be noted that in the theoretical work based on the oscillations of screening[10,11] the comparison of the theory

TABLE I. Sample characteristics: sample signature, electronic density, well width, distance d_0 from the interface to the δ layer of parent donors, electron mobility, and experimental temperature.

Sample	N_S (10^{11} cm^{-2})	Width (Å)	d_0 (nm)	μ (cm^2/V s)	T (K)
*B*20	5.0	250	25	67000	0.5
*M*422	8.2	130	11	250000	4.2
*J*508	7.8	250	11	460000	4.2
*J*514	8.55	250	10	450000	4.2

with experimental data was very preliminary. Experimentally, the observed MPL peaks do not occur at even filling factors, they never appear in the form of cusps and there is no evidence in the literature for the phase reversal of peaks for different well widths, as predicted in Ref. 12. The newer theoretical work [13,14] contains no comparison with experimental data.

In the present paper we reanalyze both experimentally and theoretically the B nonlinearities of interband energies in MPL experiments on modulation doped GaAs/$\mathrm{Ga_{0.67}Al_{0.33}As}$ quantum wells with only one conduction subband populated by electrons. We propose a transfer hypothesis assuming that the observed B nonlinearities are caused by oscillations of the 2D electron density, N_S, in a GaAs QW as the magnetic field B is varied. In other words, we treat the GaAs quantum well as an open system in contact with an outside reservoir. The density oscillations are just a result of this assumption. According to this interpretation, the $N_S(B)$ oscillations cause periodic modifications of the electron potential which results in changes of the conduction and valence band energies. A very good description of phase and amplitude of the observed B nonlinearities is obtained. In particular we find that, in agreement with experiment, the MPL peaks occur at lower magnetic fields than those corresponding to even filling factors ν, contrary to predictions of Refs. 10 and 11. In addition, no phase reversal of the MPL peaks is predicted and observed on QWs of different widths, contrary to the prediction of Ref. 12. Additional arguments in favor of the transfer interpretation are presented.

The paper is organized in the following way: In Sec. II we briefly describe our experiments and characterize our samples. In Sec. III we present the experimental data and compare them with calculations based on the transfer interpretation. In Sec. IV we discuss our results, compare them with the work of others authors, and draw conclusions. The paper concludes with a summary.

II. EXPERIMENTS AND SAMPLES

Our magnetophotoluminescence experiments were performed in the standard way: optical excitation was provided by an Argon laser, the spectra were analyzed using a monochromator-type spectrometer. The light was detected using a photomultiplier and a GaAs detector. For weak signals we must modulate optical excitation and use lock-in amplification. The samples were placed in a cryostat allowing a stable temperature. Temperatures were measured by a carbon resistor placed near the sample. An external magnetic field in the intensity range of 0–14 T was provided by a superconducting coil. A MPL spectrum was usually taken as a function of energy at a constant magnetic field. In order to determine the MPL energies when more than one optical peak was observed, we deconvoluted the experimental spectra employing Gaussian and Lorentzian forms of peaks. The measurements of transport in the dark and under illumination were performed on samples in the standard form of a double cross.

The investigated samples, grown by MBE, were asymmetric modulation-doped GaAs/$\mathrm{Ga_{0.67}Al_{0.33}As}$ single quantum wells with different well widths. The structure was δ doped in the GaAlAs barrier on one side by two planes of silicon donors, the closer being at a distance d_0 from the interface. One sample, B20, was δ doped in the well with Be acceptors at 20 Å from the interface. The sample characteristics are indicated in Table I.

In Fig. 1 we show the energy band scheme for the sample B20. The indicated subband energies and wave functions were calculated self-consistently. Using transport measurements, we checked that in the applied range of magnetic fields only the ground electric subband was populated with electrons. In Fig. 2 we give experimental MPL spectra for sample M422 as functions of frequency at fixed magnetic fields.

III. RESULTS AND INTERPRETATION

Figure 3 shows the results of our MPL experiments on sample M422. The lowest energy (D) is related to the mag-

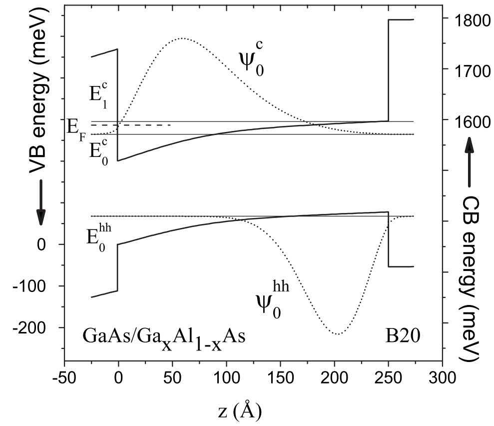

FIG. 1. Band structure of the modulation-doped GaAs/$\mathrm{Ga_{0.67}Al_{0.33}As}$ quantum well (sample B20) versus growth dimension. The calculated conduction and heavy-hole subband energies and the corresponding wave functions are shown. The position of the Fermi level is indicated.

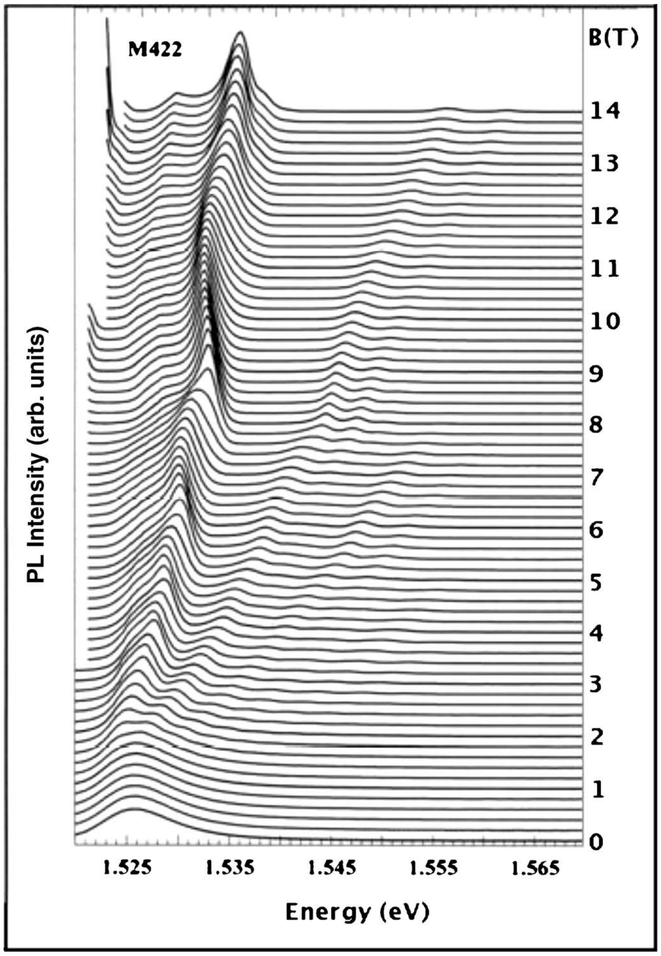

FIG. 2. Experimental MPL spectra for sample $M422$ as functions of energy at fixed magnetic fields.

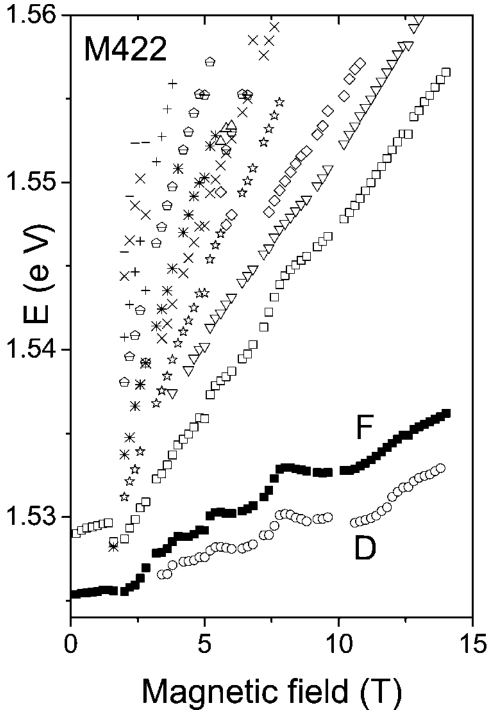

FIG. 3. Fan chart of experimental magnetophotoluminescence energies (sample $M422$) versus magnetic field. The lowest transition (D) is due to donors, the transition F (0^+-1β) is described in the text.

netodonor and we will not consider it here (see Ref. 15). We will be concerned with the energy marked F, relating to the free-electron transition from $0^+(c)$ to $1\beta(hh)$ Landau levels in the notation of Ref. 2, corresponding to the change of angular-momentum $\Delta j = -1$ (left circular polarization). The other three samples $J508$, $J514$, and $B20$ showed similar nonlinearities of the MPL transition energy for the same transition and it is this problem that we address below.

It is known by now that, in experiments using an external magnetic field B, the 2D electron density, N_S, in $\mathrm{GaAs/Ga_{0.67}Al_{0.33}As}$ QWs is not constant but undergoes oscillations as a function of B, see Refs. 16 and 17. In other words, as the magnetic field changes, the electrons are partly transferred back and forth between the GaAs QW and an outside reservoir. We propose a hypothesis that, in a structure having only one subband populated by electrons, the observed nonlinearities of MPL energies are caused by oscillations of the 2D density N_S in the GaAs well. According to our interpretation, the $N_S(B)$ oscillations cause periodic modification of the self-consistent electric potential. This, in turn, changes the conduction and valence subband energies and results in the nonlinearities of the MPL interband energies. To calculate the variations of $N_S(B)$ we take the Fermi energy E_F fixed at its value for $B=0$. This assumption is confirmed by experimental observations on samples under illumination.[18,19] Our hypothesis is equivalent to assuming that the reservoir of electron states outside the well is much larger than the number of states in the well. If this criterion is not well satisfied, the Fermi energy is fixed only partly and the density oscillations still occur but with a smaller amplitude.

Our calculations are performed in the standard way which can be described as the self-consistent Hartree approximation. Since we deal with the n-type structures, the Schrödinger and Poisson equations are solved self-consistently for the conduction subband including band's nonparabolicity. The potential energy takes into account the offset between GaAs well and GaAlAs barriers, the space charge in GaAs, the movable charge in GaAs, and the exchange and correlation energy of the electron gas in the parametrized form of Hedin and Lundqvist. The doping layer is treated as a δ function. The density of electron states in the presence of a magnetic field is taken in the form of Gaussian peaks. We take into account the fact that the width of the peaks, Γ, is also an oscillatory function of B due to the changes of screening and the resulting changes of the scattering strength.[15] We assume that $\Gamma_{\mathrm{max}} = 2$ meV when E_F is in between LLs, and $\Gamma_{\mathrm{min}} = 0.25$ meV when E_F is in the middle of a LL.

An important feature of our modeling is the exchange enhancement of the spin g factor when the Fermi energy occurs between two spin levels, so that the populations of these levels differ. The g factor enhancement is calculated according to the theory of Ando and Uemura.[20] The enhanced g value is included self-consistently in a sense that it

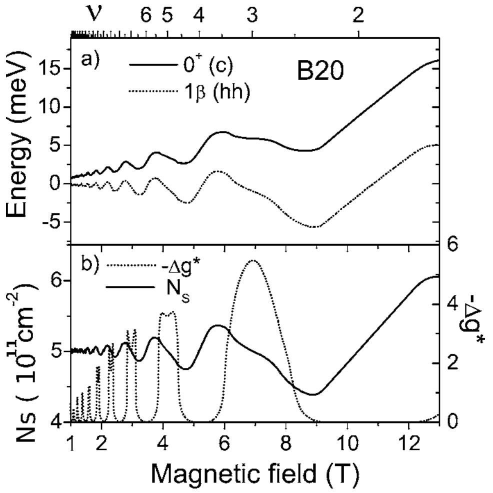

FIG. 4. Energies of the conduction level $0^+(c)$ and the heavy-hole level $1\beta(hh)$ versus magnetic field for sample *B*20. (b) Calculated electron density N_S (in 10^{11} cm^{-2} units) and exchange enhancement of the spin *g* value $-\Delta g^*$ versus magnetic field for the same sample. The corresponding filling factors ν are indicated on the upper abscissa.

both provokes and is affected by the electron density oscillations. The g^* enhancement takes place in the vicinity of the Fermi energy (see Ref. 21), but this mechanism affects the electron transfer which, via the change of the confining potential, is reflected in the behavior of all levels.

The oscillations of $N_S(B)$, the enhancement Δg^* near the Fermi level, and the energy of the conduction level E_0^+ are shown in Figs. 4(a) and 4(b). The initial parameters used for the calculations are $m^*=0.066m_0$, $g_0^*=-0.44$, and $V_B=220$ meV, where m^* and g_0^* are the effective mass and the Lande factor in GaAs, and V_B is the potential barrier for conduction electrons at the interface $Ga_{0.67}Al_{0.33}As/GaAs$. The effective mass in the $Ga_{0.67}Al_{0.33}As$ alloy is taken to be $m_0^*=0.073m_0$ and the average dielectric constant is $\epsilon_r=12.91$ for the structure.

Once the potential $V(z)$ is determined, the same potential $V(z)$ with the corresponding valence offsets is used to calculate the highest heavy-hole subband energy and the $1\beta(hh)$ LL of interest (see Fig. 1). To find the solutions for the Γ_8 degenerate valence bands described by the Luttinger model in the presence of an electric potential and a magnetic field we used the method of transition matrix described in detail in Ref. 22. As follows from Refs. 2 and 22, the heavy-hole level $1\beta(hh)$ has an almost linear dependence on *B* at magnetic fields higher than 1 T. The calculated energies of this level are shown in Fig. 4(a). It can be seen that, in agreement with our model, the oscillations of the conduction and valence energies follow the oscillations of the electron density N_S in the QW. The oscillations of $1\beta(hh)$ level are in phase with those of $0^+(c)$ level but their amplitude is somewhat larger. According to our interpretation the difference between the two energies determines the observed MPL energy. It should be mentioned that the filling factors ν, as indicated in Fig. 4(a) and all subsequent figures, correspond to real electron density N_S in the well, including the oscillations.

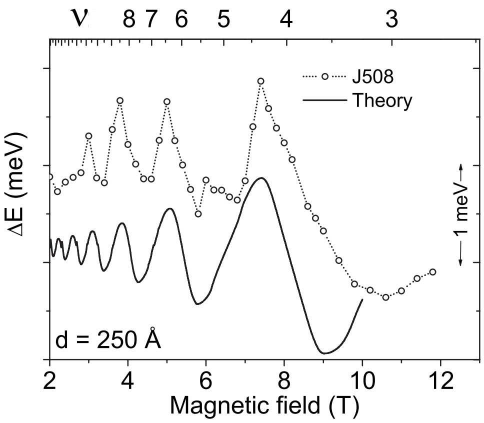

FIG. 5. Oscillatory part of the MPL energy for $0^+(c)$-$1\beta(hh)$ transition versus magnetic field (sample *J*508). The calculated curve (solid line) has been shifted down for clarity.

We find that the comparison between experimental and theoretical energies is more conclusive when the linear *B* dependence of the oscillating energies is subtracted. In Figs. 5 and 6 we compare the experimental oscillatory parts of the MPL energy for samples *J*508 and *M*422 with our calculations. As indicated in Table I, the two samples have markedly different widths but similar densities N_S. It can be seen that the experimental oscillations have almost the same amplitude in the two cases. More importantly, there is no reversal of phase due to the change of the width, contrary to the prediction of Ref. 12. On the other hand, the experimental results are in very good agreement with our calculations. In particular, our model predicts that the peaks of the MPL energies occur for magnetic fields lower than those corresponding to even filling factors ν. This is fully confirmed by the data. In our interpretation the even filling factors ν correspond to B_ν values for which the electron density $N_S(B_\nu)=N_S(B=0)$. At these B_ν values the MPL oscillations should reach their average value. This is in fact quite well confirmed experimentally. In our model, the width of QW plays no role since the equilibrium conditions relate only to the conduction subband. As a result, our model predicts no reversal of phase for different widths of QWs, in agreement with the observations. The experimental data for both samples exhibit a clear effect of the g^* enhancement at the filling factor $\nu=5$, as predicted

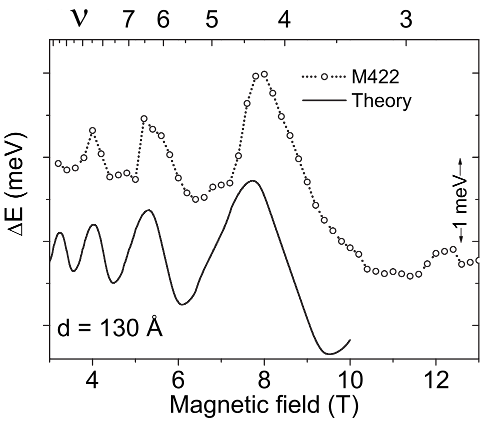

FIG. 6. The same as in Fig. 5 but for sample *M*422.

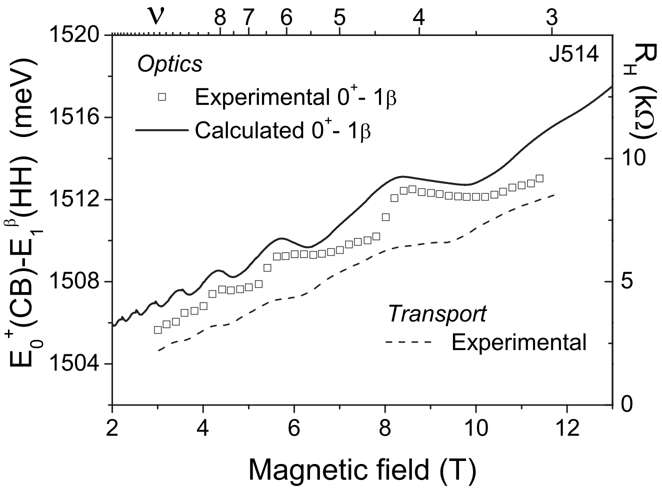

FIG. 7. MPL energy for $0^+(c)$-$1\beta(hh)$ transition versus magnetic field (sample J514). The solid line is calculated. Experimental trace of the quantum Hall effect measured on the same sample under illumination is shown for comparison.

by the theory. For sample *M*422 this seems to be the case also for $\nu=7$. On the other hand, the peaks related to odd filling factors are barely seen in the theoretical modeling. Technically, the reason is seen in Fig. 4(a) which shows that the energy peaks related to $\nu=3$ are rather flat, so that their difference comes out as an inflection point. It is not clear why the peaks related to odd ν are more pronounced in the experiment than in the calculations. On the contrary, the g enhancement at $\nu=3$ is clearly observed in the quantum Hall data, see below.

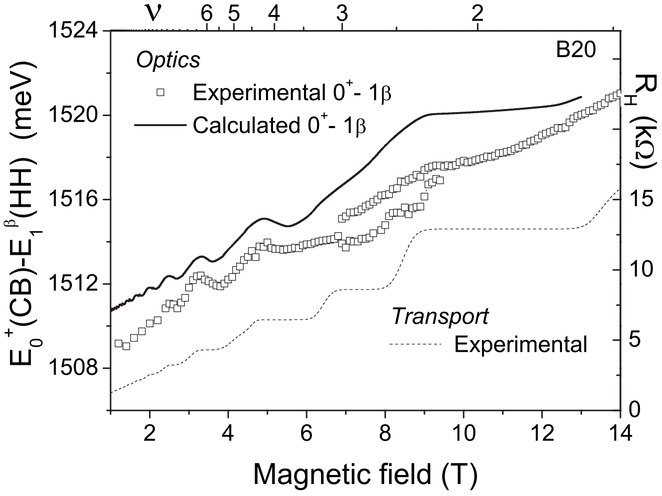

FIG. 8. The same as in Fig. 7 but for sample *B*20. The splitting of the MPL line at the filling factor $\nu \leq 3$ can be seen (see text).

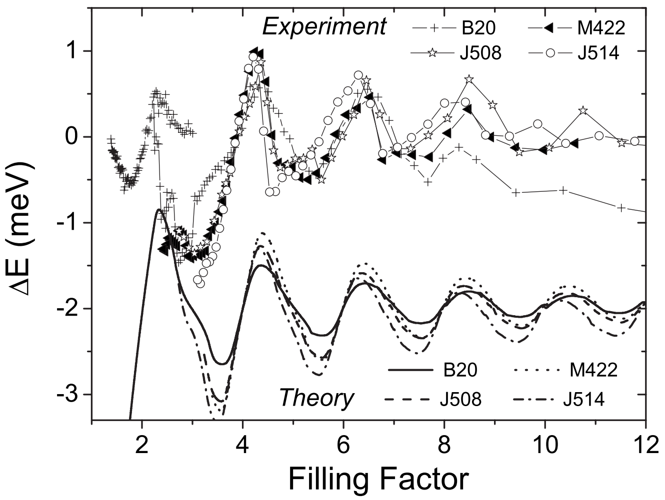

FIG. 9. Oscillatory parts of the MPL energies for four samples versus filling factor ν. The calculated curves have been shifted down for clarity.

In Figs. 7 and 8 we plot directly the MPL experimental energies for samples *J*514 and *B*20, respectively, without subtracting the linear *B* dependence. We do this in order to show in the same figures experimental traces of the quantum Hall effect (QHE) measured under illumination on the same samples. Our calculations (solid lines) again describe quite well the MPL curves and, in particular, they give very well the maxima points. The QHE traces allow us to identify without ambiguity the corresponding filling factors. It can be seen that the MPL maxima occur at lower fields than those corresponding to even filling factors. Since the middle of a QHE plateau occurs around an even filling factor, it is directly seen that the peaks of MPL occur not at even values but at higher values of ν (or lower magnetic fields). For sample *B*20 the centers of the QHE plateaux do not correspond exactly to the integer filling factors because this heterostructure has been doped in the well with Be acceptors, which makes the broadened Landau levels asymmetric. The QHE plateau for $\nu=3$ in Fig. 8 is more pronounced than it would follow from the exchange enhancement of the g^* value at this filling factor. The quality of QHE data taken on sample *B*20 is quite good in contrast to sample *J*514. This difference is not clear. The sample *B*20 has the lowest electron density in the well (see Table I) and is the only one to have been doped with acceptors in the well. The splitting of the MPL line at $\nu=3$, as seen in Fig. 8, was observed before[23] and it was attributed theoretically to a splitting of the spectral function of the final hole states.[24,14]. The origin of the splitting is a strong coupling of holes with opposite spins. We do not go into details here of this many-body effect.

In order to summarize our findings for the four samples having different electron densities, we plot the data as functions of the filling factor ν. The results are shown in the upper part of Fig. 9. In this plot all oscillations of the MPL energies exhibit a very similar behavior. The maxima occur for the same ν values and the amplitudes of oscillations decrease the same way with ν. The only "anomaly" occurs for sample *B*20 near $\nu=3$, where the MPL line splits into two, as discussed above. In the lower part of Fig. 9 we plot the corresponding theoretical curves $\Delta E(\nu)$ for the four samples. The experimental behavior is reproduced very well, both as far as the phase and the amplitude are concerned. The theoretical curves are somewhat "rounder" than the experimental ones near the maxima, this is related to the assumed width of the Landau levels.

IV. DISCUSSION

In addition to our transfer hypothesis concerning the oscillating electron density in the QW we use auxiliary assumptions which are well satisfied. Thus we consider only one conduction subband populated with electrons. We checked this assumption both theoretically and experimentally using the quantum transport data. In one of our samples ($J514$) the Fermi energy is near the first excited subband. However, we did not observe any anomalous behavior of MPL and transport compared to other samples. We emphasize that the value of the 2D electron density is not enough to decide how many electric subbands are involved. It was shown in Ref. 6 that, when dealing with a flat potential, even at a relatively low density of $N_S = 1.8 \times 10^{11}$ cm^{-2} one may deal with two populated subbands.

As to our main hypothesis concerning the electron transfer between the GaAs quantum well and the reservoir in the quantum Hall regime, it had been directly confirmed by measurements of the 2D density with the use of the cyclotron resonance.[16,17] Also, this hypothesis had been proposed in the explanation of the quantum Hall effect.[25–30] We do not go here into the interpretation of the QHE which is a much more complicated problem than the interpretation of MPL. The reason is that the dc transport involves localized electron states, mobility edges, sample edges, etc. On the other hand, in the optical phenomenon of MPL the localized and delocalized electron and hole states contribute in a similar manner. However, it is clear that also in the standard interpretation of the QHE involving the localization, the density oscillations will play a role since the actual density $N_S(B)$ determines at what magnetic fields the Fermi energy crosses the mobility edges of a given LL. In other words, $N_S(B)$ determines magnetic field positions of the quantum Hall plateaux. The reservoir hypothesis had been also considered in the interpretation of magnetization and of the thermoelectric power of 2DEG.[31] It describes both effects quite well. Kerridge *et al.*[32] demonstrated that, once a reservoir is actually *provided* by a δ layer of Si donors in the GaAlAs barrier, the charge transfer between the reservoir and the GaAs well, as modulated by a magnetic field, dramatically affects the nonlinearities of the MPL energies. We consider this experiment to be a strong argument in favor of the transfer hypothesis for the MPL nonlinearities.

Clearly, the oscillatory screening in 2D systems in the presence of a magnetic field does exist and it was experimentally observed by various authors, see, for example, Refs. 15 and 33. We include this effect when we vary phenomenologically the width of Landau levels in our modeling, as mentioned above. However, the oscillatory screening is not seen directly in MPL because, as follows from Figs. 3, 7, and 8, the complete MPL curves do not have the shape of pronounced peaks. The question arises, as to whether the oscillatory screening and correlation effects, invoked until present to explain the B nonlinearities of MPL energies, also play a role in the MPL behavior. We can say that, judging by the presented theoretical results concerning the phase and the amplitude of MPL oscillations,[10–14] the role of oscillatory screening and correlation effects in our MPL experiments is small.

Obviously, we cannot decide whether *all* doped III-V quantum wells should be treated as open systems in connection with outside reservoirs. We can only say that almost all III-V QWs exhibit B nonlinearities of MPL energies. In consequence, if our interpretation is correct, there is a strong indication that these QWs are open systems. The exact nature of the reservoir is unknown. The reservoir can be provided by the donors in a barrier which are always there if one needs high electron densities to reach the quantum Hall regime (see Ref. 28), it can also be provided by surface states of the structure. We believe that the electron transfer between the well and the reservoir is realized by means of tunneling since both optical as well as dc transport experiments are usually performed in a stationary regime. Also, it is clear that the light changes equilibrium conditions in the system which is manifested by modifications of the quantum Hall data under illumination. However, the number of photoexcited carriers is much smaller than the number of transferred electrons. All in all, our interpretation of the MPL nonlinear energies for systems with only one populated subband, as confronted with the published work on systems with more populated subbands, can be generalized by saying that in *all* systems exhibiting B nonlinearities of the MPL energies the charge transfer is at work.

V. SUMMARY

We investigated experimentally and theoretically B nonlinearities of the magneto-photoluminescence energies in asymmetric modulation doped GaAs/GaAlAs quantum wells as functions of a magnetic field. We argue that, in structures having only one electric subband populated with electrons, the MPL nonlinearities are caused not by an oscillatory renormalization of the gap due to oscillatory screening, as claimed in the literature, but by an oscillatory transfer of electrons between the GaAs well and the reservoir outside the well. The oscillatory electron density affects the potential of the well which, in turn, influences the conduction and heavy-hole subband energies. Our interpretation gives a very good description of the observed B nonlinearities in four samples of different well widths. We correctly account for the MPL maxima positions and the oscillation amplitudes. No change of the maxima into minima is predicted for increasing well widths, in agreement with the experiments. We conclude that various experimental data reported in the literature which exhibit B nonlinearities of the MPL energies in 2D systems can be explained by different kinds of charge transfer.

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