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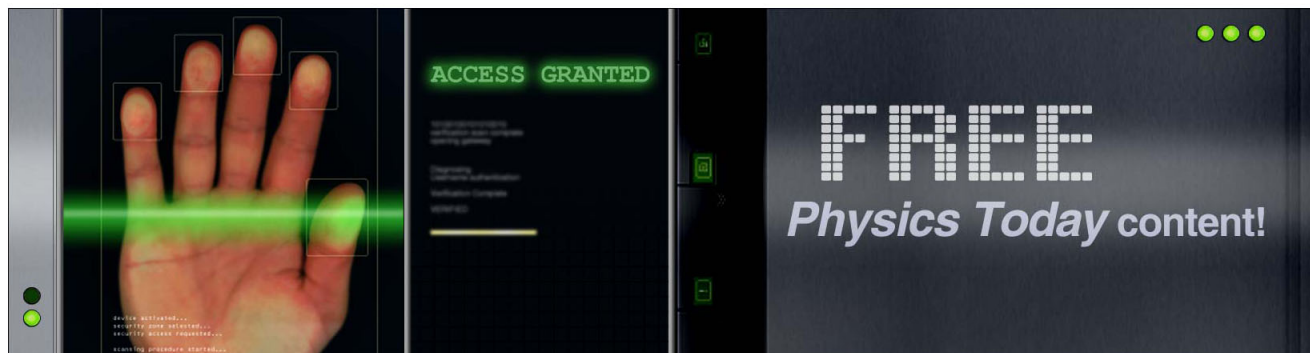
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Enhanced magneto-optical oscillations from two-dimensional hole-gases in the presence of Mn ions

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We investigated the effects of nearby Mn ions on the optical properties of two-dimensional hole-gases confined in InGaAs/GaAs quantum wells. We observed energy oscillations on both the averaged emission and the spin-splitting energies, whereas the first one presents maxima at all integer filling factors, and the second one, solely at odd-filling factors. The strength of the oscillations clearly increases with the Mn concentration. Furthermore, considering the relatively low-mobility of our structures, the oscillations are surprisingly strong and robust, persisting up to relatively high temperatures and excitation intensities. © 2011 American Institute of Physics. [doi:10.1063/1.3601477]

III–V diluted magnetic semiconductors are promising materials for spintronic applications, where the magnetic order is usually induced by the indirect exchange between localized magnetic ions mediated by free carriers.¹ For GaAs:Mn, the most studied material, the mediation is provided by holes, which is specially functional, as substitutional Mn acts as an acceptor in GaAs. The increasing density of defects generated by the introduction of Mn prevents, however, refined optical studies on GaMnAs structures. Alternative structures based on Mn δ -doping, which presented high critical temperatures,² can be used to enhance the optical and electrical response by spatially separating the Mn ions from the hole-gas confined in a quantum well. The ideal spatial separation between the Mn ions and the hole gas is a compromise reached between the magnetic properties and the quality of the hole gas.³ This system allows magneto-optical investigations of the complex valence-band hole gas and the effect of Mn on their properties.

In this letter we investigated a modulated-doped (In,Ga)As quantum-well (QW) structure with a Mn δ -doped layer at one of the GaAs barriers and a C δ -doped layer at the other barrier. Magneto-optical results from a series of samples with different Mn concentrations revealed an anomalous oscillation of the InGaAs QW emission as a function of the magnetic field, including both the peak and the spin-splitting energies. The oscillations become stronger for increasing Mn concentrations, and no oscillations could be resolved for reference samples without Mn. There are several reports concerning oscillatory behaviors on magneto-optical measurements of two-dimensional (2D) confined gases in nonmagnetic samples.^{4–11} Most of the letters present oscillations of the optical emission intensity and the peak energy, and there are some few polarization-resolved results concerning oscillations of the spin-splitting energy. The oscillations generally correlate with integer Landau level states and are usually ascribed to charge transfer^{4,5} or many-body effects.^{6–11} The majority of the results concern, however, 2D

electron-gases (2DEGs). The difficulty of observing this subtle effect on hole-gases is most likely related to their relatively lower mobilities. Magneto-optical oscillations for 2DHG structures have only been reported for specially high-quality samples with record mobilities ($\sim 4 \times 10^5$ cm²/V s), comparable to 2DEG systems.^{10,11} Surprisingly, we have observed rather strong and robust magneto-optical oscillations that persist up to rather high temperatures and laser excitation intensities, although our samples present significantly smaller mobility values ($\sim 2 \times 10^3$ cm²/V s at 77 K).

The investigated samples were grown on semi-insulating GaAs substrates using a hybrid system combining metal-organic chemical vapor deposition (MOCVD) and laser sputtering growth. First, the structure consisting of: GaAs buffer (500 nm), δ -doped C layer, GaAs spacer (10 nm), In_{0.17}Ga_{0.83}As QW (10 nm), and GaAs spacer (3 nm), was grown at 600 °C by MOCVD. The temperature was then lowered to 450 °C and a δ -doped Mn layer followed by a 15–20 nm GaAs layer were grown using laser sputtering. The nominal content of Mn (Q_{Mn}) based on the deposition rate of a thick layer was varied between 0 and 0.40 monolayers. The samples present a 2DHG with densities of $\sim 3\text{--}5 \times 10^{11}$ cm⁻². The reference sample with no Mn and the same amount of C present a slightly smaller density ($< 10^{11}$ cm⁻²). Therefore, a second reference sample was grown with additional C, resulting in a similar hole-density. We will refer to these reference samples as $Q_{\text{Mn}}=0^*$ (lower density) and $Q_{\text{Mn}}=0$ (similar density). PL measurements were performed at 2 K and magnetic fields up to 15 T along the sample growth direction using a 488 nm laser excitation ranging from 2 to 10³ W/cm². Right ($\sigma+$) and left ($\sigma-$) circularly polarized emissions were selected using appropriated polarizing components.

Figure 1 presents the magneto-optical results for two samples, without ($Q_{\text{Mn}}=0^*$) and with ($Q_{\text{Mn}}=0.40$) Mn. Typical photoluminescence (PL) spectra at 0 T are presented in the inset and the energy shift in the $\sigma+$ and $\sigma-$ polarized QW emission bands are displayed as a function of the magnetic field. The samples present remarkably different behavior.

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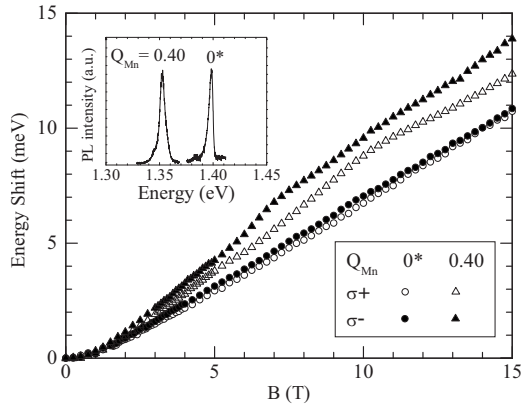


FIG. 1. Energy shift of the $\sigma+$ and $\sigma-$ QW emission peaks from a sample without ($Q_{Mn}=0^*$) and with ($Q_{Mn}=0.40$) Mn as a function of the magnetic field at 2 K and 2 W/cm². Typical PL spectra for those samples at B = 0 T are presented in the inset.

The presence of Mn implies in an increase in both, the averaged magnetic shift and the spin-splitting energy. The sample with the lowest concentration ($Q_{Mn}=0^*$) presents a quadratic magnetic field dependence, as expected for an excitonic transition, solely, for low magnetic fields. For fields larger than 2 T, the energy shift becomes essentially linear, which is characteristic of a free-carrier recombination.¹⁰ For samples with larger hole-concentrations, the binding energy of excitons should decrease due to an increased screening caused by the 2DHG, which should result in a simple free-carrierlike linear magneto-optical dependence. However, the magneto optical results for the $Q_{Mn}=0.40$ sample (Fig. 1) show a clearly nonlinear behavior with strong energy oscillations.

In order to simplify the analysis, we subtracted straight lines from the PL peak energies. They were chosen so that the subtracted energies are minimized remaining positive. The subtracted energies represent the nonlinear magneto-shift and are presented in Fig. 2 for the whole set of samples. Apart from the low magnetic field range where the magneto-shift is approximately quadratic, the samples without Mn present a negligible featureless nonlinear magneto-shift, in agreement with the results of Fig. 1. The incorporation of Mn ions clearly affects this result, giving rise to a significant nonlinear magneto-shift component that shows oscillations with the magnetic field and becomes stronger with increasing Mn concentrations.

The magnetic fields corresponding to integers filling factors of the Landau levels are indicated in Fig. 2. We observe an evident correlation between the filling factors and the magneto-shift oscillations. All Mn doped samples show a clear maximum of the nonlinear magneto-shift at $\nu=2$. For low magnetic fields, the quadratic behavior mentioned before tends to obscure possible oscillations at larger filling factors, but samples with larger Mn concentrations show stronger oscillations that can be perceived up to $\nu=5$. No oscillations could be discerned, however, for the samples without Mn doping within our experimental accuracy. Furthermore, the results are clearly different for $\sigma+$ and $\sigma-$ polarizations. The $\sigma-$ emission shows maxima only at even filling factors while the $\sigma+$ emission shows oscillations at both even and odd filling factors. Considering that the $\sigma+$ emission band appears at higher energies, this behavior results in an increased separation between the $\sigma+$ and $\sigma-$ bands at odd filling factors. This effect can be better observed by plotting the spin-

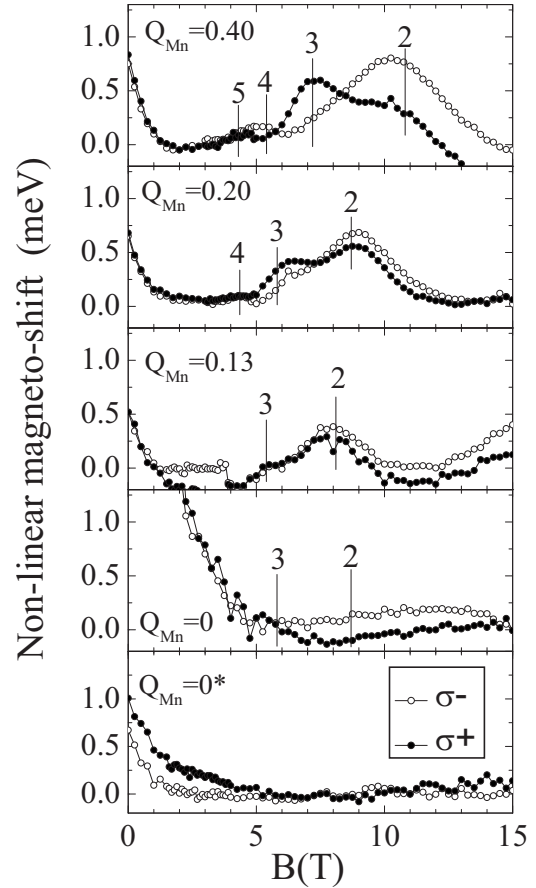


FIG. 2. Nonlinear magneto-shift as a function of the magnetic field obtained by subtracting straight lines from the $\sigma+$ and $\sigma-$ QW emission peaks for our series of samples at 2 K and 2 W/cm². The integer filling factors obtained by transport measurements are indicated.

splitting energy, which is directly obtained by subtracting the peak energies from the $\sigma+$ and $\sigma-$ bands. The results presented in Fig. 3 (continuous lines) show that the spin-splitting energy from the sample without Mn tends to saturate at ~ 0.5 meV for large magnetic fields, a behavior that has been observed and attributed to the valence-band mixing.¹² The presence of Mn ions results in the enhancement of the overall spin-splitting energy, that attains a maximum of ~ 1.5 meV at 15 T ($Q_{Mn}=0.40$), and the emergence of oscillations with maxima at odd filling factors. We also present in Fig. 3 the nonlinear magneto-shift (squares) extracted from the averaged energy of $\sigma+$ and $\sigma-$ peak energies, from which the spin-splitting energy is canceled off. Once more, we do not perceive oscillations for the sample without Mn while the Mn-doped samples show oscillations with increasing intensities and maxima at both, odd and even filling factors.

Similar results have been presented for high-quality 2DHGs with significantly higher mobility.^{10,11} Our oscillations are however stronger (~ 1.0 meV as compared to ~ 0.1 meV in Ref. 11). Furthermore, the results from Ref. 10 were performed at lower temperatures (0.3 K) and excitation intensities (2×10^{-4} W/cm²), and the magneto-oscillations from Ref. 11 completely disappeared when the temperature was increased from 0.1 to 4.2 K. In contrast, the upper panel in Fig. 3 shows that the nonlinear magneto-shift of the averaged PL emission from the $Q_{Mn}=0.40$ sample persist up to

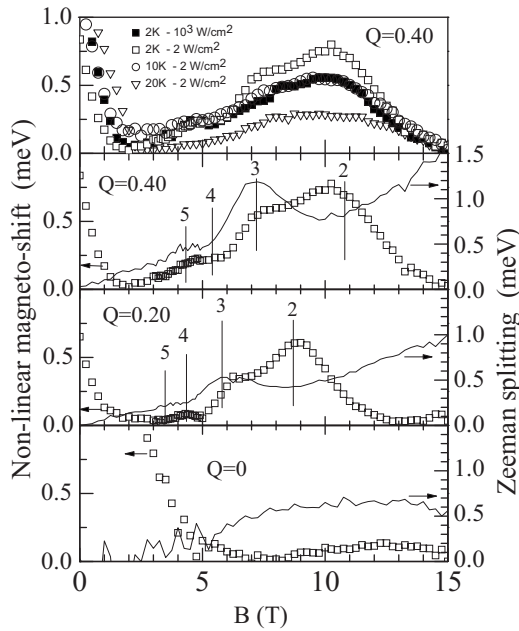


FIG. 3. Spin-splitting energy (continuous line) obtained by subtracting the $\sigma+$ and $\sigma-$ QW emission peaks and nonlinear magneto shift (squares) obtained by subtracting a straight line from the averaged energy of those peaks for the samples with $Q_{Mn}=0, 0.20$, and 0.40 , as a function of the magnetic field at 2 K and 2 W/cm 2 . Upper panel: nonlinear magneto-shift of the averaged PL emission from the $Q_{Mn}=0.40$ sample at different temperatures and excitation intensities.

20 K and an excitation intensity of $\sim 1 \times 10^3$ W/cm 2 , which indicates a rather robust effect.

Despite small differences on experimental results and theoretical models, magneto-optical oscillations of 2D gases in nonmagnetic systems have already been addressed on various letters. The most straightforward models are based on charge transfer between QW subbands. This explanation is discarded in our case, as transport measurements revealed that only one subband is occupied in our samples. Most of the magneto-optical oscillations from 2D gas systems are attributed, however, to screened many-body interactions modulated by the variation in the density of states with the magnetic field. The basic idea is that the carriers' self-energies depend on the occupation of the Landau levels, so that the energy transition presents maxima at integer filling factors.^{13,14} Spin-effects were also included in the calculations,^{15,16} but the analysis is essentially analogous. The results of those theoretical letters is that the spin-split energy oscillations are attributed to fluctuations of the Coulombic interactions, in special, variations in the exchange term due to the difference on the number of available many-particle states at odd filling factors, when one spin level is completely full and the other spin level, completely empty. As a final comment, no calculations were actually performed for 2D-hole gases, but similar results should be expected for sufficiently polarized holes.

The clear enhancement of the magneto-optical oscillations with increasing Mn concentrations revealed by our results opens a new question concerning the combined effect of the Mn ions and many-body effects. We remark that, even though the Mn ions are inserted at the GaAs barrier, a non-

negligible overlap of the QW confined holes and the Mn ions is expected due to the hole wave function penetration and the diffusion of Mn ions. The correlation between the oscillation strength and the Mn concentration and the persistence of a phenomenon traditionally attributed to many-body effects for relatively high temperatures, high excitation intensities, and low mobilities suggest a strong coupling effect between the 2DHG and the magnetic ions that acts in the way to reinforce the spin-related many-body effects in the optical transitions. The precise mechanism of these interactions remains to be investigated. The analysis of this complex system should include not only the many-body interactions among the carriers, but also the strong *sp-d* exchange interaction between holes and Mn ions.

In summary, we observed highly nonlinear polarized magneto-optical energy transitions in InGaAs/GaAs:Mn systems. The nonlinear effect is spin dependent and, clearly, associated to the presence of both the hole-gas and the magnetic impurities. They also showed remarkable resilience to relatively high temperatures and excitation intensities. At the moment, there is no theoretical model to explain those features. We hope our results will stimulate further theoretical letters on this system that may also reveal new insights concerning the hole-mediated ferromagnetic mechanism. On the other hand, the nonlinear behavior of the diamagnetic and spin-splitting energies observed for Mn:GaAs structures may be used as the basis to develop novel spintronic devices.

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