Giant Photoinduced Excitonic Faraday Rotation in $CdTe/Cd_{1-x}Mn_xTe$ Multiple Quantum Wells

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We report a giant photoinduced Faraday rotation in a $CdTe/Cd_{1-x}Mn_xTe$ multiple quantum well structure when the laser frequency is tuned close to the fundamental excitonic transition of the quantum well and saturation sets in. At the same time we observe stimulated emission and buildup of magnetization through photon spin recycling in the presence of a magnetic field which was analyzed by a time-resolved linearly polarized pump and probe technique. The magnetization buildup is also seen in the absence of the seeding magnetic field and by using a circularly polarized pump beam. [S0031-9007(97)03246-8]

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The behavior of photoinduced carriers in confined semiconductor nanostructures under the influence of static magnetic fields attracts increasing attention [1–3] that parallels the one that concerns static electric fields and at the same time contrasts with the latter. This contrast can be traced to the fundamentally different impacts that electric and magnetic fields have on the charge motion with regard to its topology and spatial extension and also with respect to the time inversion operation.

In a spatially confined one-electron system, for example, atoms, where the discrete electronic states are essentially determined by the interplay of kinetic and potential energies this contrast is most conspicuous in the appearance of Stark shifts and Zeeman splittings of the optical transitions in the presence of static electric and magnetic fields, respectively; their counterparts on the refractive indices are the static Kerr and Faraday rotation (FR) effects, respectively, which have found several applications. In spatially confined many-electron systems on the other hand, for example, semiconductor nanostructures, the electronic states are shaped through a subtle balance of more complex interactions superimposed on the bare kinetic and potential energies whose relative impact is modified differently by the static electric and magnetic fields and the corresponding optical spectrum is more complex. This is even more striking in the nonlinear optical regime where intense light beams are involved provoking population modifications and level shifts that under certain light intensities can match those provoked by the static fields. This can have important applications as well if properly controlled and in addition can be used as a probe to study certain interactions which are buried under the linear regime.

In this Letter we report the observation of a giant photoinduced FR that, in fact, is as large as the linear one [4], when the frequency is tuned across the fundamental excitonic transition of CdTe quantum wells in a $CdTe/Cd_{1-x}Mn_xTe$ multiple quantum well structure and presents an analysis of the interplay of the different interactions in the saturation regime of the excitonic transition. The key point is that by reaching the saturation regime a primary linear feature is depleted leaving other features unbalanced and thus allowing them to build up to an extent that their impact can be visible.

The sample used in this experiment was grown by molecular beam epitaxy at 280 °C under excess Cd flux. The (001) oriented structure contains ten periods of 47 Å thick wells of CdTe and 950 Å thick barriers of $Cd_{1-x}Mn_xTe$ ($x = 0.17$). These were grown on a $Cd_{1-x}Zn_yTe$ ($y = 0.22$) substrate (gap = 1.781 eV) after the deposition of a 3.9 μm thick buffer layer of $Cd_{1-x}Mn_xTe$ ($x = 0.17$). Finally, a cap layer of 1 μm of CdMnTe with the same manganese concentration was grown on the top of the structure. Below 40 K, where the measurements were performed, the substrate is transparent near the first excitonic transition of the CdTe quantum wells, so that no etching was needed. The transmission spectrum reveals a dominant oscillator strength corresponding to the $1s$ heavy hole excitonic transition with a binding energy $E_B = 23$ meV and a Bohr radius $a_B = 50$ Å. In the following discussion we shall concentrate our attention on this feature.

The sample was mounted in a magnetic cryostat with the possibility to continuously vary the magnetic field up to 5 T, and its temperature was fixed at 5 K. The FR spectra were measured by determining, as a function of the photon energy, the FR angle in a one-beam experiment using the simple technique described in Ref. [5] which provides information regarding the nonlinear regime without using a probe beam. In a second set of experiments, the FR of a probe beam is measured for different intensities of a pump beam with fixed wavelength. The laser pump and probe beams are produced through parametric generation and amplification in beta-barium-borate (BBO) crystals pumped by the third harmonic of a single pulse mode-locked Nd:YAG laser. These sources emitted nearly Fourier limited pulses ($\Delta t = 20$ ps, $\Delta E = 0.1$ meV)

tunable from 0.4 to 2.5 μm. This allowed both spectral and temporal resolution, which was not the case using femtosecond systems. Moreover, due to spatial filtering by a monomode optical fiber the beams were nearly Gaussian which allowed good reproducibility of the pump and probe experiments [6].

The giant linear excitonic Faraday rotation in $CdTe/Cd_{1-x}Mn_xTe$ multiple quantum wells can be traced to the strong spin exchange interaction between band electrons and Mn impurities [7] that also the electrons in the CdTe well experience as their wave functions penetrate into the magnetic CdMnTe barrier of finite height. Such an interaction leads to a large Zeeman splitting of the excitonic transition of the nonmagnetic CdTe well and to a subsequent giant Faraday rotation [4].

At high laser fluences, higher than 0.5 $\mu J/cm^2$, the nonlinear regime sets in and the Faraday rotation spectra undergo drastic modifications. As an example Fig. 1 shows one beam Faraday rotation and transmission spectra recorded at a magnetic field of 2 T for several laser fluences: 4.5, 32, and 200 $\mu J/cm^2$. For comparison we also show the FR spectrum for a fluence of 9 nJ/cm^2, which was checked to correspond to the linear regime. The onset of the saturation of the Faraday rotation angle is clearly visible already with a fluence of 4.5 $\mu J/cm^2$ but complete saturation throughout the spectral width of the excitonic transition is hard to achieve as attested by the factor of 40 between the fluences corresponding to spectra recorded at 200 and 4.5 $\mu J/cm^2$. Concomitant with the onset of the saturation a blueshift of 1.8 meV is also evident in Fig. 1 [8]. Their compound effect thus results in photoinduced Faraday rotations, determined by the difference between the nonlinear and linear FR, as large as the linear one, namely, $\sim 20°$ at a laser fluence of only 4.5 $\mu J/cm^2$ so that the photoinduced FR process in quantum wells can be used as a very sensitive diagnostic technique. In the case of the highest fluence of 200 $\mu J/cm^2$ the density of absorbed photons, which due to saturation is essentially frequency independent, was measured to be 12 times larger than the maximum allowed excitonic density $N_m = 1.3 \times 10^{12}$ cm^{-2}, as calculated from $N_m = 1/\pi a_B^2$ by assuming close packing of excitons.

All these features observed at a magnetic field intensity of 2 T are even more pronounced at 5 T where the σ_+ and σ_- spectral components are clearly separated. In Fig. 2 we show the FR spectra recorded in the linear regime and at a moderate laser fluence of 4.5 $\mu J/cm^2$. When the laser frequency is tuned close to the low frequency σ_+ transition one observes saturation and blueshift of the Faraday rotation angle. All these features which correspond to complicated physical mechanisms can be qualitatively accounted for in terms of the level scheme depicted in the inset of Fig. 2. Indeed because of the creation of σ_+ excitons at high laser fluence the $|-\frac{3}{2}\rangle \rightarrow |-\frac{1}{2}\rangle$ excitonic transition is saturated and, in addition, undergoes a blueshift because of many body effects related to Coulomb interaction and Pauli exclusion principle, which lead to an increase of the excitonic transition energy [9].

We also notice an increase of the absolute value of the Faraday rotation angle around 1.661 eV; this amplification of the Faraday rotation can also be seen in Fig. 1 at 1.664 eV for a pump fluence of 4.5 $\mu J/cm^2$ before saturation sets in at higher fluences. This increase of the absolute value of the Faraday rotation angle, and the difficulty to achieve saturation of the excitonic transition, can only be understood by evoking the formation of additional transitions, e.g., by exciton localization which drain the σ_+ transition [10,11]. In such a case a population inversion can occur for these states which can provide laser emission [12] which depletes the population and hampers the onset of the complete saturation. In

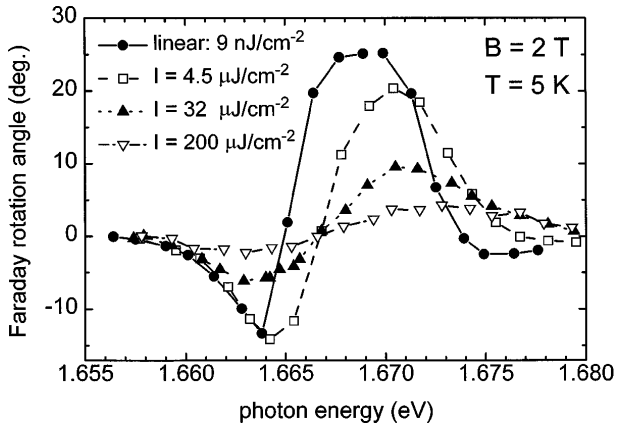

FIG. 1. Faraday rotation spectra recorded at $T = 5$ K and $B = 2$ T for different laser fluences. The angle origin is adjusted by fitting the linear FR spectrum by a two level system [4].

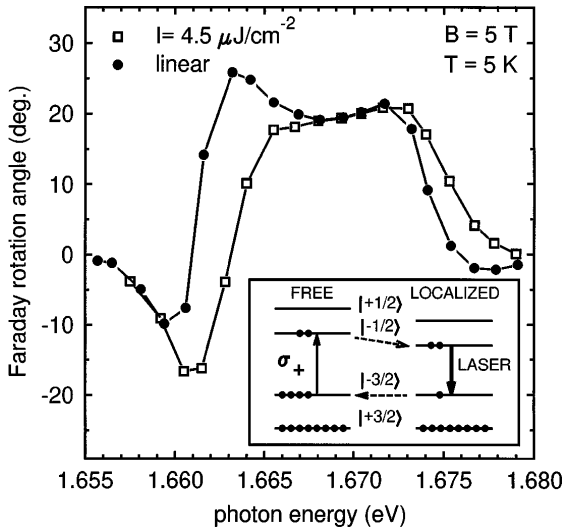

FIG. 2. Faraday rotation spectra recorded at $T = 5$ K and $B = 5$ T in the linear and saturated ($I = 4.5$ $\mu J/cm^2$) regimes. The angle origin is adjusted by fitting the linear FR spectrum by a two level system [4]. The inset shows the level scheme for conduction and valence band electrons.

our case, as in Ref. [12], due to the small thickness of the active region (10×50 Å) the laser emission did not occur in the pump beam direction but was observed in the perpendicular one where the interaction length is much longer ($\sim$0.3 mm), with a threshold of $\sim 1\ \mu\mathrm{J/cm^2}$. Moreover, the excitation of localized excitons provides an extra contribution to the Faraday rotation angle; as gain rather than absorption occurs for these red frequency shifted inverted transitions the corresponding Faraday effect reverses its sign resulting in an increase of the absolute value of the Faraday rotation angle as observed in our experiments. We have discussed here the case of gain due to localized excitons, as in Ref. [12]; however, other mechanisms providing gain on the low energy side of the excitonic line are not ruled out [13,14].

When the laser frequency is tuned close to the high frequency σ_- resonance a blueshift is visible but neither saturation nor gain occur. We ascribe this to the enhanced Zeeman shift due to magnetization transfer to the Mn^{++} system during the relaxation of excitons in the same level scheme of Fig. 2. Because of fast spin relaxation of the carriers [15], the population of the σ_- excitons is transferred to the $|+\frac{3}{2}\rangle \rightarrow |+\frac{1}{2}\rangle\sigma_+$ excitonic transition which is saturated and blueshifted instead of the $|-\frac{3}{2}\rangle \rightarrow |-\frac{1}{2}\rangle\sigma_-$ one, as it is apparent around 1.675 eV on the saturated FR spectrum of Fig. 2. In such a case, spin relaxation of electrons and holes occurs most probably due to the spin exchange interaction with those of Mn^{++} ions, the total spin being conserved [16,17]; this leads to decrease and increase of the magnetization for electrons and hole relaxations, respectively. As the spin charge is 3 times larger for holes than for electrons, there is a net increase of the magnetization [18]. On a complete optical pumping cycle, from a σ^+ photon to a σ^- one, a magnetization $2\mu_B$ is transferred to the Mn system, and the Zeeman shift correspondingly increases. Moreover, as in the case when the laser frequency is near the σ_+ resonance, the free σ_+ excitons can transfer their energy to localized excitons which provide laser emission and the large densities of absorbed photons also observed experimentally in this spectral region. The combined effects of spin relaxation of excitons and photon recycling by laser emission make the magnetization sufficiently large to explain the blueshift of the σ_- excitonic transition observed experimentally [1.5 meV in reasonable agreement with the Zeeman shift estimated for the density of absorbed photons ($2.5 \times 10^{12}/\mathrm{cm^2}$ per quantum well)]. Finally, as the incident laser frequency is far from the lasing transition resonance, it is not amplified and there is no increase of the Faraday rotation angle as also experimentally observed (see Fig. 2).

This interpretation is also supported by time-resolved pump and probe experiments performed by using two independently tunable picosecond parametric sources. The pump beam modifies the Faraday rotation experienced by the probe beam. Figure 3 shows, in solid lines, the FR spectrum of the probe beam in the presence of the pump beam for three different pump-probe delays of 0, 40, and

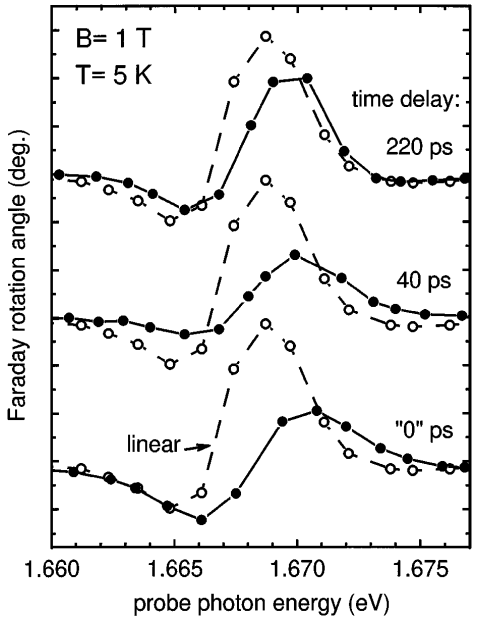

FIG. 3. Time-resolved pump and probe photoinduced Faraday rotation spectra recorded at $T = 5$ K and $B = 1$ T (closed circles). The linear Faraday rotation spectra (open circles) are shown for comparison.

220 ps at a magnetic field of 1 T. For convenience, each one of these spectra is compared to the linear FR spectrum obtained at the same field (broken lines in Fig. 3). For this experiment the pump beam had a moderate fluence ($4.5\ \mu\mathrm{J/cm^2}$) and its frequency (1.67 eV) was chosen at the center of the excitonic line with no magnetic field present so that σ_- and σ_+ transitions were equally excited by the linearly polarized pump pulse. For zero delay the FRS of the probe beam looks like that obtained in the one-beam experiment (see Fig. 1) with the blueshifts of the σ_+ and σ_- sides of the spectrum and the saturation and gain on the σ_+ side only. For a delay of 40 ps, which is twice the pulse duration, the σ_- side of the spectrum is almost unchanged while on the σ_+ side a strong saturation of the Faraday rotation angle appears, but no gain. This is consistent with our previous analysis since laser emission from localized excitons, which due to the high excitation regime (10^{27}–$10^{29}\ \mathrm{cm^{-3}\,s^{-1}}$) occurs only during the pump pulse duration, annihilates the population inversion and gain as soon as there is no more time overlap between the probe and pump pulses.

At the longer pump-probe delay of 220 ps, the σ_- side of the spectrum recovered almost completely in accordance with our model since magnetization relaxation is expected to be very fast at the high 17% Mn^{++} concentration of our sample [19,20]. It is also interesting to note that the σ_+ side of the spectrum partly recovered in 220 ps, indicating a complete recovery in less than one nanosecond which is not the case when considering the interband Faraday rotation effect already observed in bulk samples where much longer lifetimes were measured [21]. This short recovery time for the excitonic photoinduced Faraday

rotation observed in quantum wells could be of importance for fast optical switching.

We also point out that the observed saturation behavior is not related to a heating of our sample since at the 10 Hz repetition rate of our laser the average power was low, about 2 μW, and Faraday rotation spectra recorded at negative delays were identical to the linear ones.

The buildup of the magnetization reported above was evidenced in the presence of an applied static magnetic field which differentiates the left and right circularly polarized components of the linearly polarized pump beam. Indeed, in a series of pump and probe experiments without static magnetic field no rotation of the probe beam polarization was observed with linearly polarized pump beam. However, the effect can be initiated by a circularly polarized pump beam in the absence of any "seeding" magnetic field. A strong probe polarization rotation is then induced, 6° at 1.672 eV as shown in the photoinduced FR spectrum of Fig. 4, with circularly polarized pump beam of 1.671 eV and an energy of about 10 $\mu J/cm^2$. Furthermore, as expected, this strong rotation, which amounts to 30% of the maximum rotation obtained with a 1 T magnetic field and linearly polarized pump beam, reverses its sign when we reverse the circular polarization of the pump beam. Clearly this optically induced magnetization can be exploited for the study of the nonlinear magneto-optical properties of these semimagnetic semiconductors at room temperature.

In conclusion, we have furnished evidence of a giant photoinduced Faraday rotation in CdTe/CdMnTe multiple quantum wells close to the excitonic transition of the CdTe well which for moderate magnetic fields and laser fluences matches the linear Faraday rotation. Taking into account the thickness of the multiple quantum well this amounts to an effective photoinduced Verdet constant of 7×10^8 deg T^{-1} m^{-1} for pump energies of only 50 $fJ/\mu m^2$. This effect can be used as a very sensitive tool for diagnostics of electron spin interactions in confined nanostructures [1]. In this respect the saturation regime we observed here by depleting the primary energy channel permits enhancement of the effect of other contributions and favors stimulated processes like laser emission and the buildup of an effective magnetization by photon spin recycling, an induced magnetization also observed with no applied magnetic field when using circularly polarized pump beams.

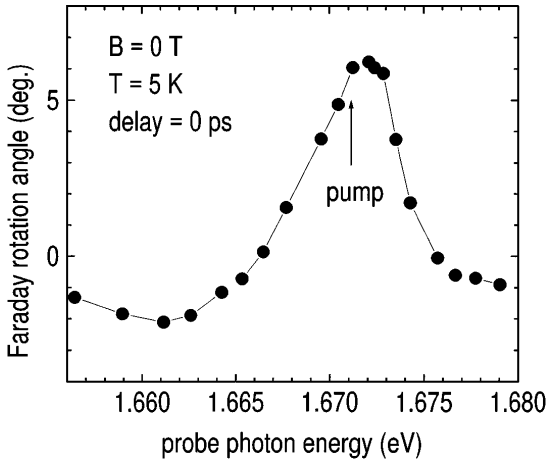

FIG. 4. Photoinduced Faraday spectrum recorded at $T = 5$ K and $B = 0$ T for a circularly polarized pump beam.

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