Zeeman mapping of probability densities in square quantum wells using magnetic probes

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We use a method to probe experimentally the probability density of carriers confined in semiconductor quantum structures. The exciton Zeeman splitting in quantum wells containing a single, ultranarrow magnetic layer is studied depending on the layer position. In particular, a system consisting of a 1/4 monolayer MnTe embedded at varying positions in nonmagnetic CdTe/CdMgTe quantum wells is investigated. The *sp-d* exchange interaction results in a drastic increase of the Zeeman splitting, which, because of the strongly localized nature of this interaction, sensitively depends on the position of the MnTe submonolayer in the quantum well. For various interband transitions we show that the dependence of the exciton Zeeman splitting on the position of the magnetic layer directly maps the probability density of free *holes* in the growth direction.

Band-gap engineering and molecular-beam epitaxial growth allow us to tailor eigenstates and wave functions of free carriers in semiconductor heterostructures. Although there are many spectroscopic tools probing eigenstates in such structures, there are only very few experimental techniques directly measuring electron wave functions or probability densities (PD's). On metal surfaces, scanning tunneling microscopy (STM) has been used to fabricate nanostructures and to probe the wave functions of the bound states.[1,2] While STM probes the lateral extension of the wave function, in semiconductor heterostructures quantum confinement is usually achieved in growth (vertical) direction. In these structures, the vertical rather than lateral dependence of the PD is of interest. It can be obtained experimentally, e.g., by photoemission experiments using synchrotoron radiation[3] as has been recently achieved in the case of metallic samples or, in the case of semiconductor heterostructures, by resonant magnetotunneling between one-dimensional quantum confined states, where the Fourier transform of the final-state wave function can be deduced.[4] In a rectangular quantum well, the differences between transition energies induced by insertion of a precisely controlled potential perturbation were optically measured and used to extract the differences of the PD at the location of the potential spike.[5] Similarly, in a parabolic quantum well with an inserted thin barrier the vertical variation of the PD was determined by transport experiments measuring differences between energy states.[6]

In this paper, we map PD's in heterostructures by exploring the exciton spin (Zeeman) splitting in quantum wells with inserted ultrathin probe layers containing paramagnetic Mn^{2+} ions. The observed spin splitting is proportional to the PD at the position of the local probe. The inclusion of magnetic submonolayers results in a strong increase of the Zeeman splitting,[7] due to spin-spin exchange interactions occurring between *s*-like conduction-band electrons and *p*-like valence band holes and the *d* electrons of the Mn^{2+} ions (*sp-d* interactions).[8] Therefore, the Zeeman splitting can be easily measured by magneto-optical spectroscopy.

Recently, the Zeeman splitting has been used to pinpoint the localization of wave functions in coupled triple and quintuple quantum-well samples containing semimagnetic quantum wells as magnetic probes.[9] In these multiquantum-well samples it is important that the width of the magnetic wells, which are probing the wave functions, are equal to that of the nonmagnetic wells, in order to obtain strongly coupled states. This condition is limiting the vertical resolution of the ''PD map.'' In contrast, in this paper a much higher resolution is obtained since magnetic probes with thicknesses in the order of 1 ML (monolayer) are used, hardly influencing the PD's under investigation.

The *sp-d* interaction is described by the exchange Hamiltonian[10,11]

$$H_{ex}=\sum_{\vec{R}_i} J^{sp-d}(\vec{r}-\vec{R}_i)\vec{S}_i\vec{\sigma}, \tag{1}$$

where $\vec{S}_i$ and $\vec{\sigma}$ are the spin operators of the Mn^{2+} ions and of the band electron, J^{sp-d} is the electron (hole)-ion $s(p)$-d exchange coupling constant, and $\vec{r}$ and $\vec{R}_i$ are the coordinates of the band electron and of the magnetic ions, respectively. Due to the localized character of the $3d$ electrons of the Mn ions, the function J is strongly peaked around $\vec{R}_i$ and vanishes quickly away from this point. Therefore, $J^{sp-d}(\vec{r}-\vec{R}_i)$ can be approximated by a delta function $J^{sp-d}(\vec{r}-\vec{R}_i)=A\Omega_0\delta(\vec{r}-\vec{R}_i)$, where A is the exchange constant[12] and Ω_0 the volume of the unit cell. The electron wave function can be written as:[12]

$$\Psi(\vec{r};\vec{S}_1,\ldots,\vec{S}_N)=\Psi(\vec{r};\{\vec{S}_i\})=\psi_e(\vec{r};\{\vec{S}_i\})\Phi(\{\vec{S}_i\}). \tag{2}$$

The symbol $\{\vec{S}_i\}$ denotes the set of quantum numbers required to label the states accessible to the system of local moments. The exchange term leads to an increased spin splitting of the conduction band, given by:[12]

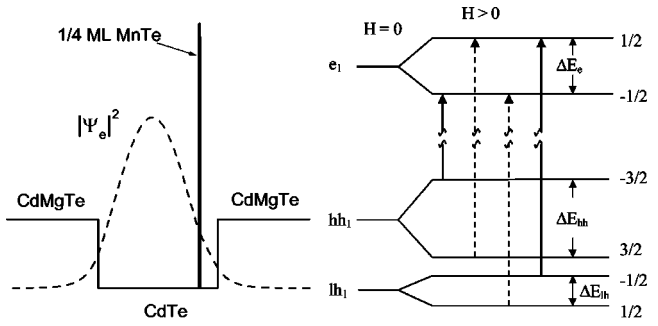

FIG. 1. Left side: Conduction band of sample $S5$ together with the electron probability density calculated for the ground state e_1. Right side: Schematic representation of optical transitions allowed for circularly polarized light with left (full lines) and right (dashed lines) helicity in Faraday geometry.

$$\Delta E_e = \sum_i \langle \Phi | S_i^z | \Phi \rangle A \Omega_0 |\psi_e(R_i)|^2. \tag{3}$$

Since in our structures the electrons are confined in growth direction, z, one can write the wave function as: $\psi_e(r=R_i)=\varphi_e(X_i,Y_i)\xi_e(Z_i)$. Moreover, all the magnetic ions are distributed within a single submonolayer, so that $Z_i=Z_{Mn}$ for all magnetic ions. Therefore, ΔE_e is proportional to $|\xi_e(Z_{Mn})|^2$ and the z dependence of the PD can be explored by measuring ΔE_e as function of Z_{Mn}. We note that in Eq. (3) the macroscopic average of S^z vanishes in the absence of an external magnetic field. Expressions analogous to Eq. (3) can be obtained for the splitting of the $\sigma=1/2$ light holes $[\Delta E_{lh}(Z_{Mn})]$ and of the $\sigma=3/2$ heavy holes $[\Delta E_{hh}(Z_{Mn})]$.

We investigate a series of five single quantum-well structures ($S1$ to $S5$). The wells consist of 20 MLs of CdTe (approximately 6.5 nm thick) between 200 nm thick $Cd_{0.75}Mg_{0.25}Te$ barriers. Each quantum well contains a narrow, magnetic MnTe barrier with a Mn amount equivalent to 1/4 ML coverage. In sample $S1$ the MnTe layer is inserted after the third ML of CdTe, while in $S2$ to $S5$ the magnetic probes are embedded after the seventh, tenth, thirteenth, and seventeenth ML of CdTe, respectively. As an example, the conduction-band edge of $S5$ is shown in Fig. 1(a). It shows, in addition, the calculated PD of the electron ground state ($|\xi_{e1}(Z_{Mn})|^2$), which is only slightly affected by the insertion of the ultrathin MnTe barrier (details of this calculation are described below). The result shows that the probe practically does not disturb the quantity that should be measured.

All samples were grown by molecular-beam epitaxy on a (001)-oriented GaAs substrate at a substrate temperature of 280°C. The layer-by-layer growth was controlled *in-situ* by observing reflection high-energy electron diffraction oscillations at each stage of the nucleation. In samples grown under identical conditions, the thickness of a nominal 1 ML thick MnTe barrier was determined by magneto-optical spectroscopy in combination with rapid thermal annealing to be equivalent to 1.25 ML's,[13] enabling the fabrication of the desired narrow magnetic layers. Taking into account this intermixing length the Mn concentration in the center of the 1/4 ML amounts to about 20%. This value is close to the concentration where the maximum of the Zeeman splitting is obtained in bulk $Cd_{1-x}Mn_xTe$.[14]

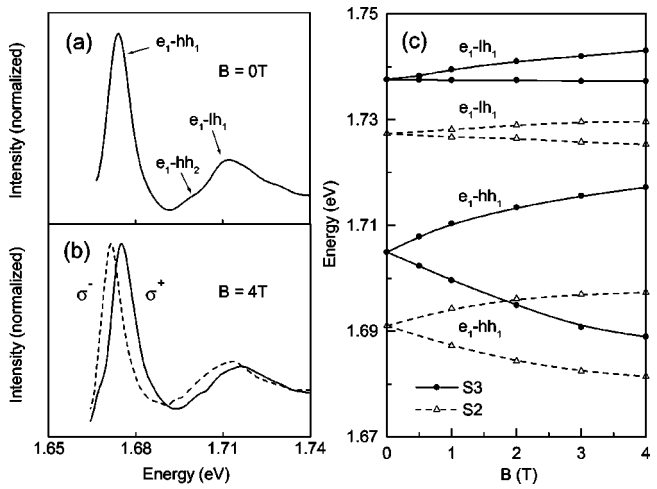

FIG. 2. PLE spectrum of sample $S5$ detected at the PL maximum, without (a) and with applied magnetic field (b). (c) Interband Zeeman splitting of the e_1-hh_1 and the e_1-lh_1 transition for sample 2 and 3 measured at 1.7 K.

The magneto-optical measurements were performed in a Faraday-configuration where optical transitions are allowed for circularly polarized light with positive or negative helicity.[10] We performed polarization dependent photoluminescence (PL) and PL excitation (PLE) experiments in magnetic fields up to 4 T. The samples were mounted in the center of a split coil magnet and immersed in superfluid helium at a temperature of 1.7 K. For excitation a tunable Coherent CR 599 dye laser was used, operating in the wavelength range between 620 nm and 800 nm. For all optical transitions probed by PLE, the Zeeman splitting is given by the sum of the spin splitting of the involved electron state (ΔE_e) and that of the hole state (ΔE_h), as schematically shown in Fig. 1(b).

At a temperature of 1.7 K, the PL spectrum of each sample consists of a single, Gaussian shaped line with a full width at half maximum of about 6 meV. The PLE spectrum of each sample is recorded at the maximum of the corresponding PL spectrum. Without magnetic field, the PLE spectrum of $S5$, shown in Fig. 2(a), exhibits a narrow line at 1.665 eV due to the e_1-hh_1 exciton transition. We observe two additional features, one peak at 1.712 eV due to the lowest light hole transition (e_1-lh_1) (Ref. 15) and a faint shoulder at 1.698 eV. Previously, this shoulder was attributed to an excited heavy-hole transition.[15]

The PLE spectra for samples $S1$–$S4$ are similar to that in Fig. 2(a), except that the optical transition energies continuously shift to higher energies when the MnTe barrier is moved from one side of the quantum well towards its center. The maximum shift exceeds 30 meV. In principle, the energy shifts of these optical transitions can be used to determine the z dependence of the PD of free carriers, as demonstrated in Ref. 5. Such a procedure requires, however, several assumptions, e.g., about exciton binding energies, and only approximate values for the PD's of the lowest electronic states can be obtained due to the finite number of transitions observed.[5] Our results are, in contrast, not affected by the exciton binding energy, since the Zeeman splitting is deter-

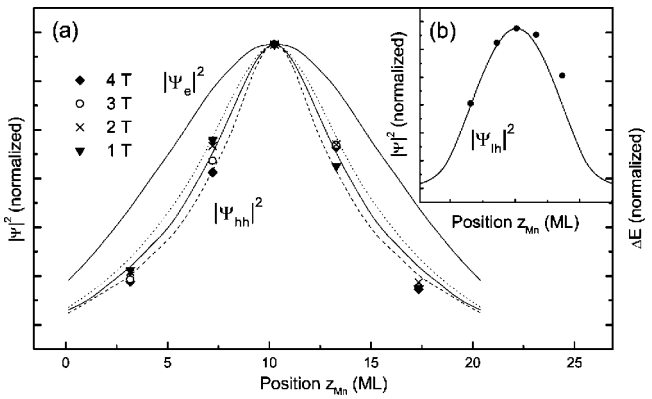

FIG. 3. (a) Probability density (PD) of the heavy-hole ground state calculated for various valence band offsets [0.25 (dashed), 0.33 (solid), and 0.45 (dotted)] and of the electron ground state (valence band offset 0.33). The symbols display the experimentally observed interband Zeeman splitting of the e_1-hh_1 transition. (b) PD of the light-hole ground state compared with the Zeeman splitting of the e_1-lh_1 transition.

mined from energy differences between measured transitions so that excitonic contributions to the transition energies are canceled out.

With applied magnetic field, each optical transition splits into two distinct lines. This is demonstrated in Fig. 2(b) for sample $S5$, where spectra obtained at 4T under σ^+ and σ^- polarized PL excitation are shown. The magnetic-field dependent transition energies, summarized in Fig. 2(c) for samples $S2$ and $S3$, are deduced from numerical fits of the individual PLE spectra. For $S3$ at 4 T, a maximum exciton Zeeman splitting of the e_1-hh_1 transition of 29 meV is found, while it amounts to 16 meV for $S2$ and $S4$, and only to 5 meV for $S1$ and $S5$. Furthermore, the observed spin splitting of the e_1-lh_1 transition is found in all samples to be 3 to 5 times smaller than that of the e_1-hh_1 transition.

Figure 3(a) shows the dependence of the Zeeman splitting of the e_1-hh_1 transition on the position of the MnTe layer in the quantum well. This dependence is compared with the electron and hole PDs, $|\xi(Z_{Mn})|^2$, calculated for the ground state of our quantum wells perturbed at the position Z_{Mn} by a quarter ML MnTe. For this calculation we used the following parameters: $E_g(\mathrm{Cd}_{1-x}\mathrm{Mg}_x\mathrm{Te})=(1.606+1.654x)$ eV,[16] $E_g(\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te})=(1.606+1.592x)$ eV,[10] $m_e^*=0.096m_0$, $m_{hh}^*=0.63m_0$ (m_0 is the free electron mass),[17,18] and we took three values for the valence band offset, 0.25, 0.33, and 0.45, in the range given in the literature.[19] Independent of the chosen valence-band offset, we found an excellent agreement between the Z_{Mn} dependence of the experimental Zeeman splitting and the PD of the heavy-hole ground state [Fig. 3(a)], for all applied fields. In contrast, the z dependence of the electron PD clearly deviates from the experimental $\Delta E(Z_{Mn})$ confirming that the exciton Zeeman splitting is dominated by the heavy-hole spin splitting. This agrees with the situation in bulk $\mathrm{Cd}_{1-x}\mathrm{Mn}_x\mathrm{Te}$, where the heavy-hole splitting is four times larger than that of the electrons.[10] Therefore, by exploring $\Delta E(Z_{Mn})$ we map out the z dependence of the PD mainly of holes in contrast to the previous work, where wave functions of electrons have been probed.[4–6]

Figure 3(b) shows that the experimentally found dependence of the Zeeman splitting of the e_1-lh_1 transition on Z_{Mn} matches the evaluated PD of the light-hole ground state. The latter nearly shows the same z dependence as the PD of the e_1 state. Therefore, a comparison of the experimental spin splitting with the calculated PD does not allow to decide, whether the spin splitting of the electrons or that of the light holes dominates the Zeeman splitting of the e_1-lh_1 transition. In principle, this ambiguity could be cleared, by separately measuring the spin splitting of the conduction band and that of the valence band, e.g., by performing spin-flip Raman scattering experiments.

In summary, we have probed, by magnetic interactions, the vertical distribution of the probability density of carriers confined in a semiconductor quantum structure. The interband Zeeman splitting of a set of quantum well samples containing narrow MnTe barriers was investigated by photoluminescence excitation experiments. We found a strong dependence of the spin splitting on the position of the magnetic layers in the quantum well. This observation implicitly confirms the strongly localized nature of the *sp-d* exchange interaction and justifies to approximate the exchange constant J^{sp-d} by a δ function. Furthermore, the Z_{Mn} dependence of the experimentally determined Zeeman splitting agrees with the calculated probability density for the heavy-hole ground state and the light-hole ground state. Thus, incorporation of magnetic submonolayers allows us to map directly the probability density distributions of *holes* confined in quantum structures, with a vertical resolution of approximately 0.4 nm, by measuring the interband Zeeman splitting. Since the narrow magnetic probes hardly influence the probability densities under investigation, this technique can be applied not only to square quantum wells but also to more complicated kinds of multiheterosturctures.

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