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Interwell exciton relaxation in semimagnetic asymmetric double quantum wells

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Possibility of a magnetic field control of spectral and polarization characteristics of exciton recombination is examined in Cd(Mg,Mn)Te-based asymmetric double quantum wells. At low fields, exciton transition in semimagnetic well is higher in energy than that in nonmagnetic well and interwell exciton relaxation is fast. In contrast, when the energy order of the exciton transitions reverses at high fields, unexpectedly slow relaxation of σ^--polarized excitons from nonmagnetic well to the σ^+-polarized ground state in the semimagnetic well is observed. Strong dependence of the total circular polarization degree on the heavy-light hole splitting Δ_{hh-lh} in the nonmagnetic well is found and attributed to the spin dependent interwell tunneling controlled by exciton spin relaxation. Such a slowing down of relaxation allows separation of oppositely spin-polarized excitons in adjacent wells.

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New field of solid state physics – spintronics, related with spin manipulation, e.g. spin alignment and transfer, has attracted much attention during last years [1]. Possibility to control energy level position and spatial localization of carriers in artificial semiconductor nanostructures has invoked a great practical interest to such nanodevices as to perspective components for future electronics. On the other hand, invariable fundamental interest to this field remains due to rich diversity of spin-related properties of the nanostructures [1].

Diluted magnetic semiconductor (DMS) nanostructures exhibit effective carrier relaxation to the ground state with characteristic times in the picosecond range [2]. Great advantage of the DMS structures is that a giant Zeeman effect in such materials as CdMnTe makes possible continuous tuning of the band gap and exciton energies by external magnetic field, due to strong $s,p-d$ exchange interaction between free carriers and localized d-states of magnetic ions [3]. This allows to vary the interwell coupling in a double quantum well (QW) structure after growing, when a barrier width L_B is fixed.

In this work we present evidence of a strong influence of spin and energy exciton relaxation on the spectral and polarization characteristics of exciton recombination in asymmetric double QWs (ADQWs) with one nonmagnetic (NM) QW and another DMS QW in continuous waves (cw) experiments in magnetic field. One should note, that the basic idea of the separation of excitons with opposite spin polarization traces back to the idea of spin superlattice with alternating DMS and NM layers [4, 5]. In presented work we found that the slowing down of spin relaxation of photoexcited σ^--polarized excitons in the NM QW gives rise to suppression of exciton tunneling to the σ^+-polarized ground state in the DMS QW. As a result, one can separate excitons with opposite spins in different QWs.

CdMgTe/CdTe/CdMgTe/CdTe/CdMnTe undoped ADQWs were grown by molecular beam epitaxy on a thick CdTe buffer deposited on (001)-oriented CdZnTe substrate. As grown structures have external 25-nm wide NM ($Cd_{0.8}Mg_{0.2}Te$) and DMS ($Cd_{0.8}Mn_{0.2}Te$) barriers and two pure 6-nm wide CdTe QWs, separated by NM $Cd_{0.8}Mg_{0.2}Te$ barrier with L_B= 3, 4 and 6 nm. Samples were covered by a thin SiO_2 cap layer of 100 nm and subjected to a rapid temperature annealing (RTA) once (L_B= 3 and 6 nm) or two times (L_B= 4 nm) at 400 °C for one minute to promote diffusion of Mn and Mg atoms from the barriers into the QWs. As we reported previously [6], diffusion is strongly enhanced below the SiO_2 mask as compared to non-covered areas and results in the increase of the QW band-gap E_g below the mask up to 0.3 eV depending on processing parameters. RTA technique allows to vary E_g in controllable way with a good optical quality [6]. QW located between two CdMgTe barriers incorporates only Mg atoms after RTA and is referred to as NM

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QW whereas that with nearby DMS barrier contains both Mg and Mn atoms and is referred to as DMS QW. Photoluminescence (PL) was excited by a cw tunable dye-laser and measured in superfluid ^{4}He ($T \approx 1.5$ K) in a cryostat with superconducting magnet. Circularly polarized ($\sigma^{\pm}$) excitation laser and PL signal beams were formed with quarter-wave plates and polarizer. PL and PL excitation (PLE) spectra were recorded with a double-stage 0.82 m monochromator and CCD camera.

Figure 1 shows polarized magneto-PL cw spectra of the investigated ADQWs in Faraday geometry, excited by σ^--polarized laser at ~100 meV above the exciton transitions but below the barrier band gap. The spectra in Fig.1 display broad lines (~7 meV) characteristic for

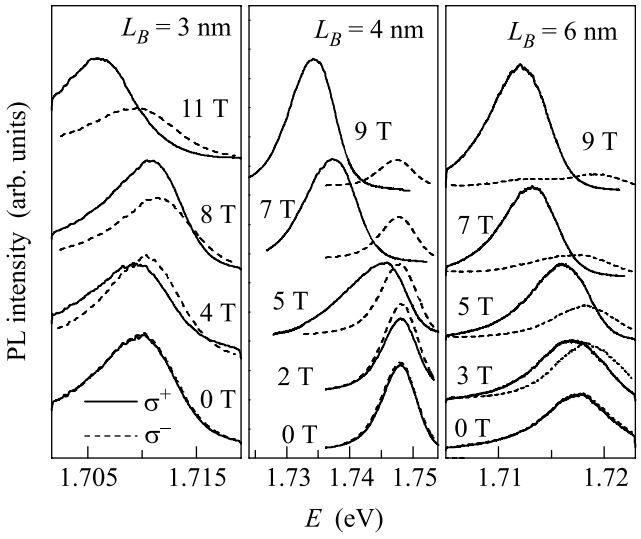

Fig.1. Magneto-PL spectra in Faraday geometry of ADQWs with L_B = 3, 4 and 6 nm. Solid lines – σ^+ polarization, open lines – σ^-. Intrawell excitation is σ^-- polarized and ~ 100 meV above exciton transition energies

ternary II-IV materials. In magnetic field B, the spectral bands split into two σ^+- and σ^--polarized components corresponding to $J = +1$ and $J = -1$ states of bright excitons [1].

In low magnetic fields, spectral positions and intensities in both polarizations are nearly B-independent in all samples, which is characteristic to NM QWs. Small excess in intensity of σ^--polarized band reflects almost complete spin relaxation of photoexcited $J = -1$ excitons. At higher magnetic fields, at some particular field value $B_C \approx 6$, 3 and 2.5 T for samples with L_B = 3, 4 and 6 nm correspondingly, a red shift of the σ^+-polarized band increases strongly while its intensity I^+ increases gradually. Such kind of behavior is characteristic to excitons in DMS QWs [3]. The higher energy σ^--polarized component, in contrast, displays a very weak shift in the whole magnetic field range. Its intensity I^- decreases with B in all structures with the stronger decrease for bigger L_B. Figure 2 summarizes magnetic field dependencies of circular polarization degree $P_C = (I^+ - I^-)/(I^+ + I^-)$. Spectral bands splitting at high fields strongly exceeds thermal energy kT indicating non-thermal distribution between $J = +1$ and $J = -1$ exciton states. Thus, spin relaxation in the studied ADQWs at $B > B_C$ takes much longer time than that in a single DMS QW.

PLE measurements have shown that all pronounced exciton transitions are intrawell ones. The lowest exciton transition in all samples weakly depends on magnetic field at low B in both polarizations and thus we ascribe it to as spatially localized in the NM QW. The first excited exciton level quickly decreases energy with magnetic field in σ^+ excitation polarization and is ascribed to the DMS QW. Due to a strong disorder in the studied ADQWs, no evidence of Landau level formation was found till $B = 12$ T. As in the case of PL, at some particular B, close to B_C, exciton transition in the DMS QW becomes lower in energy than that in the NM QW. PL and PLE data for the ground exciton transition correspond each other with the value of stokes shift of about 7–10 meV in agreement with large PL linewidth.

Note that energy sequence of the $J = +1$ and $J = -1$ exciton states in magnetic field is opposite in NM and DMS CdTe-based QWs. In case of the DMS QWs, the main contribution in the magnetic field splitting of electron and hole states is provided by the strong sp–d exchange interaction with localized Mn^{2+} magnetic moments, which results in a huge positive g_e and negative g_{hh} effective g-factors of an electron and a heavy hole (hh) respectively [3]. At $B > 0$ ground exciton transition in the DMS QW is σ^+-polarized and corresponds to optical transition between the upmost state with $J_z = -3/2$ moment projection in a valence band Γ_8 and the lowest state with $S_z = -1/2$ spin projection in conductivity band Γ_6 [3]. Usually, a quick relaxation of carriers from σ^--polarized excited exciton state, induced by the strong sp–d exchange [2], gives rise to observation of only single σ^+-polarized ground exciton transition in PL. Opposite to DMS QWs, in nonmagnetic CdTe wells g_{hh} is small and positive while g_e is negative [7]. As a result, the lowest bright exciton state is σ^--polarized in the NM QW.

Stronger exciton relaxation at high B, observed as stronger increase of polarization degree P_C in ADQWs with more thick barriers (Fig.2), does not allow to relate

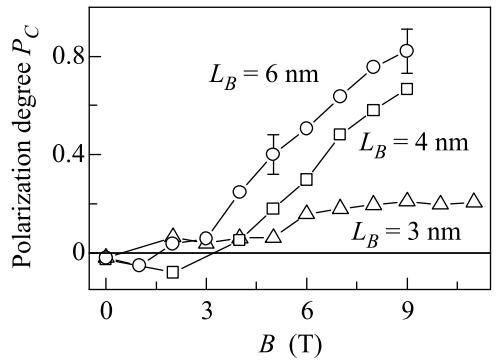

Fig.2. Magnetic field dependence of circular polarization degree P_C for spectra in Fig.1. Bars show experimental error

interwell relaxation directly with tunneling. To consider exciton relaxation in the investigated ADQWs we have calculated single-particle energy levels. CdMnTe and CdMgTe band parameters are well known at present [3, 8]. For simplicity, we assumed rectangular conductive and valence band profiles in the normal to QWs plane direction. The calculated band alignment for the structure with L_B= 4 nm at $B = 0$ and 6 T are presented in Fig.3. Atomic contents of diffused atoms Mn($\approx$ 5.0%)

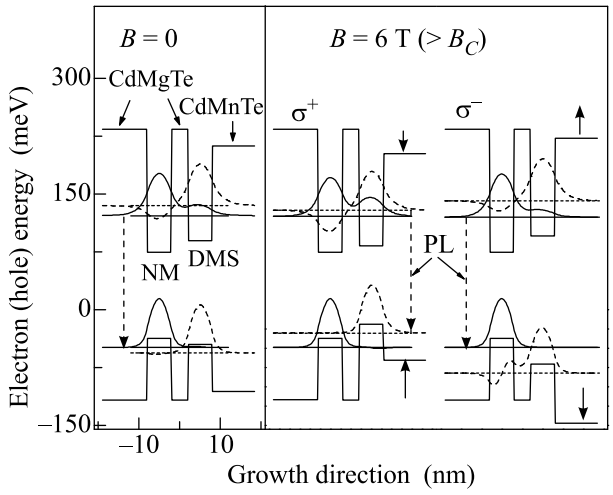

Fig.3. Calculated potential profiles in ADQW growth direction for $S_z = -1/2$ and $J_z = -3/2$ states (σ^+ label) and $S_z = +1/2$ and $J_z = +3/2$ (σ^- label) in sample with L_B=4 nm at $B = 0$ and $B = 6$ T. Electron and hole wave functions in the NM QW (solid line) and DMS QW (dashed line) are shifted according to state energy. Short arrows show direction of barriers shift with magnetic field, dashed long arrows – PL transitions

and Mg($\approx$ 5.3%) in the DMS and NM QWs, respectively, after RTA treatment have been estimated from the fit of calculated exciton energies to measured ones by PLE.

The giant Zeeman effect in the DMS QW and barrier dramatically decreases band energies for σ^+ configuration (Fig.3). hh levels of the DMS and NM QWs reverse their sequence with magnetic field at $\approx$ 1 T for all ADQWs, according to the calculations. One can see also that for the $J = -1$ exciton in the NM QW (σ^- label) both single-particle and exciton relaxation to the DMS QW are energy forbidden. Only spin-flip of the hh or of the photocreated $J = -1$ exciton in the NM QW followed by tunneling to the DMS QW can explain reduction of σ^--polarized PL, observed in the experiment at high B. Importance of spin relaxation for charge-transfer in nanostructures was found also in Ref. [9] where strong depolarization with the increase of excitation energy was observed. The experimental findings were discussed in terms of spin relaxation of hot excitons and hot carriers during intraband energy and momentum relaxation [9].

Note that single-particle spin-flip of electrons and holes in the NM QW via very effective sp–d exchange mechanism is hindered. The heavy hole cannot relax its spin via p–d exchange as a consequence of low tunnelling transparency of the barrier even at L_B=3 nm. Electron, corresponding to the σ^--polarized exciton transition in the NM QW, also cannot relax spin due to s–d exchange despite of finite penetration to the DMS QW because its state ($S_z = +1/2$) is the ground state of the NM QW. Spin relaxation of electrons via D'yakonov-Perel's spin-flip mechanism is strongly reduced at high magnetic field down to ns-time scale [10].

The exciton spin-flip, driven by a long-range exchange interaction, is known as the main mechanism of exciton spin relaxation in QWs for close to resonant photoexcitation [11]. Another strong mechanism is the hole spin-flip which is extremely quick in bulk semiconductors due to hh-lh degeneracy of the valence band [1]. Size quantization and strain effects in QWs lift the degeneracy and reduce spin relaxation with the increase of hh-lh level splitting Δ_{hh-lh}. At finite in-plane wavevector $\mathbf{k}$, the valence band states are mixed and hh spin relaxation is allowed [12]. Theoretical analysis has shown that at high $\mathbf{k}$, corresponding to in-plane kinetic energy $\geq$ 10 meV, hh spin-flip time can be as fast as a few picoseconds for ionized impurities or alloy fluctuations scattering [12] which should be the case in the studied ADQWs after RTA.

In the studied experimental conditions, carriers with large $\mathbf{k}$ are created. Relaxation of nonresonantly photoexcited carriers occurs in two main stages: first, after ultrafast subpicosecond energy relaxation by LO-phonon emission, excitons with a large center-of-mass $\mathbf{k}$

are formed during several tens picoseconds [13]. Then, for hundreds of picosecond, excitons lose their excess energy through acoustic phonon emission. Spin-flip rate via long-range exchange mechanism is increased at high $\mathbf{k}$ as compare to that for thermalized excitons at the bottom of exciton branch [11, 14]. So, high energy excitons relax spin during hot energy relaxation within time less than several tens ps – typical long-range exchange spin-flip time at near to resonant conditions [11]. hh with large $\mathbf{k}$ can also quickly relax its spin via valence band mixing. At $B > B_C$, after any spin-flip process, photocreated in the NM QW exciton with $J = -1$ can effectively tunnel and relax to the ground state in the DMS QW. As a result, spatial separation of oppositely spin-polarized excitons takes place. Thus, it is strong spin relaxation during hot energy relaxation of nonresonantly photoexcited carriers that plays major role in interwell exciton transfer in investigated ADQWs with magnetic field dependent potential profile.

Considered hh spin-flip mechanism is based on the valence band mixing: it is stronger for smaller values of Δ_{hh-lh} [12]. Δ_{hh-lh} amount to $\approx$ 34, 19 and 15 meV for the NM QW in samples with L_B= 3, 4 and 6 nm respectively, as measured by PLE. This trend exactly follows our experimental findings: stronger decrease of PL intensity in σ^- polarization takes place in structures with smaller Δ_{hh-lh}. The observed correlation points to the main contribution of the hh spin-flip because exciton spin-flip via long-range exchange interaction depends on Δ_{hh-lh} only in higher orders of theoretical expressions [11].

In summary, we have shown that by tailoring structure design in Cd(Mn,Mg)Te-based ADQWs and band parameters such as hh-lh splitting, in the first order, one can strongly influence on the intrawell spin-flip and subsequent interwell carrier relaxation in spin-dependent potential of semimagnetic ADQWs. The strong dependence of interwell exciton separation on the Δ_{hh-lh} value, found in experiment, elucidates major role of the heavy hole spin-flip mechanism due to valence band-mixing rather than exciton spin relaxation via long-range exchange mechanism at nonresonant photoexcitation. By increasing the Δ_{hh-lh} value it is possible to suppress interwell relaxation and accumulate excitons with opposite spin polarization at the excited exciton level in the nonmagnetic QW. Thus, basing on general principles of spin relaxation in semiconductor nanostructures, one can control spectral and polarization characteristics of the DMS ADQWs.

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