Heating of the Mn spin system by photoexcited holes in type-II (Zn,Mn)Se/(Be,Mn)Te quantum wells

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The efficiency of the Mn-spin system heating under pulsed laser excitation is studied in diluted magnetic semiconductor heterostructures Zn0.99Mn0.01Se/Be0.93Mn0.07Te with type-II band alignment by means of time-resolved photoluminescence and pump-probe reflectivity. An essential role in the heating is played by multiple spin-flip scatterings of a hole with localized spins of Mn2+ ions. The efficiency of the spin and energy transfer from photoexcited holes to Mn ions of the Zn0.99Mn0.01Se layer considerably depends on the hole lifetime in this layer. This lifetime can be limited by the hole relaxation into the Be0.93Mn0.07Te layers and is strongly sensitive to the excitation power and Zn0.99Mn0.01Se layer thickness. These dependences allow us to determine a characteristic time of about 20 ps for the spin and energy transfer from photoexcited holes to the Mn spin system.

1 Introduction Diluted magnetic semiconductors (DMS) have attracted great attention in the last decades as model structures for spin electronics applications, as they combine electronic semiconductor properties with a strong enhancement of spin-dependent phenomena [1, 2]. In this respect, the incorporation of magnetic ions into II–VI host material plays a key role. Strong exchange interaction of the localized magnetic moments with the conduction-band electrons and/or valence-band holes gives rise to giant magneto-optical effects [3–5].

A particular example is the exciton giant Zeeman splitting, from which one can obtain in an optical way information about the polarization of the spin system of magnetic Mn2+ ions, its spin temperature and its dynamical response on external perturbations [6–8]. This has been used to investigate in DMS heterostructures the dynamics of spin-lattice relaxation in the Mn spin system and its interaction with free carriers and phonons, both being in equilibrium or non-equilibrium.

The spin-lattice relaxation of the excited Mn2+ ions into their equilibrium with the phonon bath strongly depends on the concentration of Mn ions [9, 10]. Isolated Mn2+ ions do not interact with the phonon system in II–VI semiconductors like, e.g., (Zn,Mn)Se and (Cd,Mn)Te. However, an increase of Mn concentration leads to formation of Mn clusters having fast spin relaxation. Spin diffusion from single Mn ions to such clusters can considerably accelerate their spin-lattice relaxation [11]. It has been demonstrated that the spin diffusion plays an important role in the magnetization relaxation dynamics, especially for DMS heterostructures with a non-uniform Mn distribution [11–13]. In particular, the non-uniform Mn distribution has been realized in heteromagnetic Zn0.99Mn0.01Se/Be0.93Mn0.07Te quantum wells (QWs) with type-II band alignment [11, 13].
Photogenerated or electrically injected free carriers with excess kinetic energy can cause heating of the Mn spin system by converting their kinetic energy to the increase of the Mn spin temperature. In an external magnetic field, which induces Zeeman splitting of the Mn spin states, exchange carrier scattering on the localized Mn spins provides simultaneous spin and energy transfer into the Mn spin system. Two ways (direct and indirect) for the spin and energy transfer from the hot carriers to the Mn spin system have been found experimentally in DMS heterostructures \[14–16\]. An indirect transfer is mediated by phonons emitted by the hot carriers and absorbed by the Mn ions; the latter stage is controlled by spin-lattice relaxation processes. The direct way is governed by fast carrier exchange scattering on the localized Mn spins and is characterized by very short transfer times in the picosecond range. It was shown that the direct way is relevant for (Zn,Mn)Se based heterostructures \[14, 15\].

The requirement of the simultaneous spin and energy transfer for the carrier exchange scattering on a localized Mn spin in an external magnetic field is of key importance for the considered problem of the heating efficiency. It means that a hot carrier even having sufficient kinetic energy cannot exchange it with a Mn spin, if their spins are not properly oriented. As a result, the number of interaction processes for a hot carrier is controlled by the ratio of the carrier cooling time, which can also be limited by carrier recombination, and the carrier-spin relaxation time provided by all other mechanisms except the exchange scattering \[6\]. Accordingly, a hot electron in II–VI DMS QWs can typically flip only one Mn spin. While for a hole, with its strong spin–orbit mechanism and the carrier-spin relaxation time provided by all other mechanisms except the exchange scattering \[6\], a hot carrier is controlled by the ratio of the carrier cooling time, which can also be limited by carrier recombination, and the carrier-spin relaxation time provided by all other mechanisms except the exchange scattering \[6\]. Accordingly, a hot electron in II–VI DMS QWs can typically flip only one Mn spin. While for a hole, with its strong spin–orbit interaction and fast spin relaxation time, multiple angular momentum transfer to the Mn spin system is possible. Such a multiple spin transfer from each spin-polarized hole has been demonstrated experimentally for (Zn,Mn)Se/(Zn,Be)Se QWs with type-I band alignment \[17\]. The suggested model proposes the dominant role of the photoexcited holes in the direct heating of the Mn spin system.

(Zn,Mn)Se/(Be,Mn)Te heterostructures with type-II band alignment offer a remarkable opportunity to control the hole lifetime in the (Zn,Mn)Se layers either by structure parameters, i.e., thickness of the (Zn,Mn)Se layers \[18\], or by excitation density \[19, 20\], which controllably varies band bending \[21\]. The point is that the potential minimum of the valence band in such structures belongs to the (Be,Mn)Te layers, and the hole, which is photogenerated in the (Zn,Mn)Se layers can relax into the (Be,Mn)Te layers, see Fig. 1a. The hole relaxation time depends on the above-mentioned parameters and can vary from \(< 1 \) to \(> 100 \) ps \[20\]. Thus, it can considerably limit the lifetime of the hot holes in the (Zn,Mn)Se layers. We will explore this property of the type-II heterostructures to provide an in-depth view into the Mn heating effects by the hot carriers. In this paper, we study the relationship between the lifetime of hot holes and the efficiency of heating of the Mn spin system in Zn\(_{0.99}\)Mn\(_{0.01}\)/Se layers of Zn\(_{0.99}\)Mn\(_{0.01}\)/Se/Be\(_{0.93}\)Mn\(_{0.07}\)/Te heterostructures with different layer thicknesses under different levels of optical excitation. The experimental findings highlight the dominant role of multiple hole spin-flip processes and allow us by means of the hot hole lifetime to estimate the time of spin and energy transfer from photogenerated holes to the Mn spin system being characteristic for (Zn,Mn)Se based DMS heterostructures.

### 2 Experimental

We studied two Zn\(_{0.99}\)Mn\(_{0.01}\)/Se/Be\(_{0.93}\)Mn\(_{0.07}\)/Te undoped multi-QW structures with type-II band alignment and different layer thicknesses, denoted as 20/10 and 10/5 nm. The samples were grown by molecular-beam epitaxy on (100)-oriented GaAs substrates. They contain ten periods of alternating Zn\(_{0.99}\)Mn\(_{0.01}\)/Se layers with a thickness \(d_{qw} = 20(10)\) nm and 10(5)-nm-thick Be\(_{0.93}\)Mn\(_{0.07}\)/Te layers, respectively.

The band structure of a type-II (Zn,Mn)Se/(Be,Mn)Te heterostructure at low excitation power (flat bands) is schematically shown in Fig. 1a. At liquid helium
temperatures, the band gap of (Zn,Mn)Se with a Mn concentration of few percent is approximately 2.8 eV, and that of (Be,Mn)Te is about 4.5 eV. The localization potentials amount to ≥2.5 eV for electrons in the (Zn,Mn)Se layers and ≈0.9 eV for holes in the (Be,Mn)Te layers. In our experimental conditions, the laser light is absorbed by the (Zn,Mn)Se layers only. The photogeneration of electrons and holes in the (Zn,Mn)Se layers is followed by a relaxation of the holes within picoseconds to the energetically lower valence-band edge of the (Be,Mn)Te layers, whereas the electrons remain in (Zn,Mn)Se. This spatial carrier separation is characteristic of a type-II band alignment.

The spatially direct (D) and indirect (ID) optical transitions are indicated in Fig. 1a by arrows. In photoluminescence (PL) spectra, they appear as lines at about 2.8 and 1.9 eV at low excitation power, respectively. Normalized PL spectra at two different excitation powers are shown in Fig. 1b for the 20/10 nm sample. It is clearly seen that the spectral position and the shape of the indirect emission line strongly depend on the excitation density. The spatial separation of the photoexcited carriers in the (Zn,Mn)Se and (Be,Mn)Te layers results in strong internal electric fields, which lead to strong band bending [21]. The spectral shift of the indirect emission line and its changes during and after the laser pulse can be used to estimate the concentration of the spatially separated carriers, both electrons and holes whose concentrations are equal to each other [22]. Since in the studied structure the hole scattering to the (Be,Mn)Te layers is very efficient, the concentration of the spatially separated carriers corresponds well to the concentration of the photogenerated carriers. In the following, we will refer to it as the photo-carrier concentration.

In Ref. [22], it has been shown for non-magnetic ZnSe/BeTe structures that the band bending also causes the formation of metastable above-barrier hole states [19], which considerably increase the time $\tau_{\text{rel}}$ of the hole relaxation process from ZnSe to BeTe layers. Much stronger localization of the above-barrier hole states in samples with thicker ZnSe layers leads to strong dependence of $\tau_{\text{rel}}$ on the width of the respective layer as well [18]. All these effects determine the observed variations of the hole relaxation times for ZnSe/BeTe-based structures in the wide range from shorter than 1 ps up to longer than 100 ps [20].

For our magneto-optical experiments, we used a split-coil superconducting solenoid generating magnetic fields up to 10 T. Each sample, cooled down to a temperature of $T = 1.8$ K, was exposed to a magnetic field applied along the sample growth axis (Faraday geometry). Since the magnetization of the Mn ions is directly proportional to the giant Zeeman shift of the excitons, it can be followed by the spectral position of the exciton line [10]. The magnetization dynamics was measured by time-resolved pump-probe magneto-reflectivity. Pump pulses with a frequency of 1 kHz and 5 ns duration of the third harmonic (3.49 eV) of a Q-switched Nd:YVO$_4$ laser were used for heating the Mn spin system. Such a low frequency was chosen to assure that the excited Mn spin system was completely cooled down to the bath temperature between consecutive laser pulses. Dynamics of the heavy-hole exciton resonance in the reflectivity spectrum was measured and associated with the magnetization dynamics of the Mn spin system.

A reflectivity spectrum was detected under pulsed illumination by the light emitted from the dye Coumarin 120 in the spectral range between 2.70 and 2.95 eV. The dye was pumped by a second laser with photon energy of 3.49 eV and 5 ns pulse duration and the dye pulse repeats the shape of the excitation pulse with a very short delay, that is not relevant for our experiment. A gated charge-coupled-device (CCD) camera with temporal resolution of ≥2 ns, connected to a 0.5-m monochromator was used for detection. Both pulsed lasers and the gated CCD were synchronized via a digital pulse delay generator, which also provided the time delay for the probe dye-light. The advantage of the reflectivity against PL is that it allows to measure the average Mn spin temperature in the illuminated sample area, while regions with low Mn spin temperature more strongly contribute to the PL [23, 24].

The lifetime of photoexcited holes in the Zn$_{0.99}$Mn$_{0.01}$Se layers and its dependence on the carrier density were measured by time-resolved PL [19]. The second harmonic (≈3.0 eV) of a Ti:sapphire laser was used for PL excitation. It emitted 80 fs pulses at a frequency of 76 MHz with a maximum power on the sample surface of 200 μJ cm$^{-2}$ corresponding to the generation of an electron–hole pair density up to $2 \times 10^{13}$ cm$^{-2}$. The picosecond kinetics of the direct PL line was measured by a streak camera combined with a 0.5-m monochromator; the time resolution was 6 ps and could be improved by deconvolution down to about 2 ps.

3 Results and discussion In the inset of Fig. 2, reflectivity spectra of the 10/5 nm sample with $d_{\text{ex}} = 10$ nm are shown for $B = 0$, 0.3, and 3 T. The spectra are unpolarized;
Accordingly, they contain resonances of both \( \sigma^+ \) and \( \sigma^- \) polarizations. At \( B = 3 \text{T} \) two strongly pronounced resonances at 2.797 and 2.832 eV are seen. They correspond to the direct optical transitions. The one at lower energy stems from the \(-1/2, +3/2\) exciton giving rise to \( \sigma^+ \) circularly polarized light, while the energetically higher resonance has \( \sigma^- \) polarization [4].

The giant Zeeman shift \( \Delta E_X = E_X(B) - E_X(0) \) of the hh-exciton resonance energy \( E_X \) is shown in Fig. 2. For both samples studied, the shifts are very similar, e.g., at \( B = 3 \text{T} \) they are \(-17.0\) and \(-17.5\) meV for the 20/10 and 10/5 nm structure, respectively. It indicates that the static magnetization of the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers is very weakly influenced by the presence of Mn ions with higher concentration in the adjacent \( \text{Be}_{0.93}\text{Mn}_{0.07}\text{Te} \) layers [11] and also that the wave functions of electron and above-barrier hole states are strongly localized within the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers [20].

Pulsed laser excitation may induce heating of the Mn spin system, as it has been demonstrated for, e.g., \((\text{Zn,Mn})\text{Se})\) based QWs with type-I band alignment [14], which results in a decrease of the exciton giant Zeeman shift. The dependence of \( \Delta E_X \) on the time delay \( t_d \) relative to the pump pulse is shown in Fig. 3 for both studied structures at \( B = 3 \text{T} \) and pump density of 33 kW cm\(^{-2}\). One can clearly see that the giant Zeeman shift is reduced mainly during the laser pulse action (shown by dotted line), i.e., during the presence of hot photogenerated carriers in the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers.

For time delays \( t_d \) exceeding 40 ns and extending to few hundreds of nanoseconds, \( \Delta E_X(t_d) \) does not change considerably. The reduction of the giant Zeeman shift by \( \delta E(n) = E_X(n) - E_X(n \approx 0) = 10 \text{ meV} \) for the photo-carrier concentration \( n \approx 9 \times 10^{12} \text{ cm}^{-2} \) achieved under the pump laser density of \( P_{\text{pump}} = 33 \text{ kW cm}^{-2} \) indicates that the Mn spin system is heated to a spin temperature of \( T_{\text{se}} \approx 9 \text{ K} \) [6]. It should be noted that the Mn spin temperature is measured after the pump pulse at time delays, where the concentration of the spatially separated carriers (electrons in \((\text{Zn,Mn})\text{Se})\) and holes in \((\text{Be,Mn})\text{Te})\) layers is already small and does not exceed \( 10^{12} \text{ cm}^{-2} \) even for the highest excitation density used in our experiments [21, 22]. Therefore, the exciton resonances are well pronounced in the reflectivity spectra and can be used for the evaluation of the Mn spin temperature.

The further slow relaxation of the magnetization (not shown here) has been studied in detail for these samples in Ref. [11]. The magnetization relaxation times of \( 1.7 \mu s \) for the 10/5 nm and \( 6 \mu s \) for the 20/10 nm sample have been found. They are contributed by a very long spin-lattice relaxation in the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers and by spin diffusion in the Mn spin system removing spin excitations into the \( \text{Be}_{0.93}\text{Mn}_{0.07}\text{Te} \) layers.

Now, we turn to the main part of the paper where the Mn heating is compared for the two studied samples for different concentrations of photogenerated carriers in the range \( 10^9 \text{ cm}^{-2} \leq n \leq 1.15 \times 10^{13} \text{ cm}^{-2} \). This concentration has been evaluated by the method presented in Refs. [22, 25]. The evaluation of the photo-carrier concentration \( n \) is based on the estimation of the number of photons/cm\(^2\) from the pump laser power. Moreover, the absorption coefficient \( 4 \times 10^4 \text{ cm}^{-1} \) of the studied structure [26] and average lifetime of the photoexcited carriers are taken into account. At high pump powers, \( n \approx n_0 \approx 10^{12} \text{ cm}^{-2} \), the Mn heating is very small and the \( \delta E \) value is practically negligible.

In a broad concentration range, \( 10^9 \text{ cm}^{-2} < n < 8 \times 10^{12} \text{ cm}^{-2} \), \( \delta E \) in the 20/10 nm sample (closed squares) exceeds that in the 10/5 nm sample (open squares) by a few meV. It evidences that the Mn heating is more efficient in the sample with thicker \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers. The difference in the heating becomes most pronounced at \( n \approx 2 \times 10^{12} \text{ cm}^{-2} \), where \( \delta E \) is about \( 3.6 \text{ meV} \) corresponding to a Mn-spin temperature overheating \( \Delta T_{\text{se}} \approx 2.3 \text{ K} \) and \( 1.4 \text{ meV} \) \( (\Delta T_{\text{se}} \approx 0.8 \text{ K}) \) for the 20/10 and 10/5 nm structure, respectively. At very high concentrations, \( n \geq n_0 = 8 \times 10^{12} \text{ cm}^{-2} \), \( \delta E \) coincides in both samples. The largest \( \delta E \) value of about \( 12 \text{ meV} \) given at \( n = 1.15 \times 10^{13} \text{ cm}^{-2} \) corresponds to a spin temperature \( T_{\text{se}} \approx 15 \text{ K} \).

The different heating efficiencies found for same carrier concentrations in the two studied structures cannot be explained by the heating due to photo-electrons. First, the electrons are confined in the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \) layers and their concentration in these layers is the same in both structures and is limited by recombination only. Second, as we have discussed above, each electron can provide only one exchange scattering process with a Mn ion and, thus, the electron contribution to the Mn heating is considerably smaller than that of the holes with their multiple spin transfer [17]. Therefore, the different heating efficiencies are related to the Mn heating by the hot photo-holes, whose lifetime in the \( \text{Zn}_{0.99}\text{Mn}_{0.01}\text{Se} \)
layers depends strongly on the layer thickness and the band bending controlled by the pump power density.

The hole lifetimes in the Zn_{0.99}Mn_{0.01}Se layers are extracted from single-exponential fits to the picosecond PL decays of the direct-in-space exciton PL line. In Fig. 5a and b, the temporal behavior of the excition PL intensity is shown for both samples with photo-carrier densities being close to $n_{0}$ and 1 order of magnitude smaller. A strong difference in the decay behavior and, accordingly, hole lifetimes for both samples at the same photo-carrier density can be clearly seen. Note, the recombination time for spatially separated electrons and holes, whose concentration controls the renormalized band potential, is considerably longer than the direct recombination in the (Zn,Mn)Se layers [22, 25].

In Fig. 4b, the hole lifetime $\tau$ for both samples is demonstrated as a function of the photo-carrier density $n$. With increasing carrier density, $\tau$ changes from 16 ps up to about 100 ps in the 20/10 nm sample and from $\leq 2$ ps (our time resolution) to about 30 ps in the 10/5 nm sample. The strong increase in the hole lifetime for the 20/10 nm sample at $n > 1 \times 10^{12}$ cm$^{-2}$ in comparison to that of the other sample corresponds to a much larger Mn heating efficiency, as indicated in Fig. 4a. Due to the longer lifetime of the hot holes in the 20-nm-thick Zn_{0.99}Mn_{0.01}Se layers, the holes on average more strongly contribute to the Mn heating. At carrier densities $n < n_{0} \approx 8 \times 10^{12}$ cm$^{-2}$ the hole lifetime in the 20-nm-thick Zn_{0.99}Mn_{0.01}Se layers is much longer than in the sample with 10-nm-thick layers. Here, the Mn heating is more efficient in the 20/10 nm sample emphasizing the dominant role of the hot holes in the exchange scattering processes with the localized Mn spins. At high carrier densities, $n \geq n_{0}$, the hole lifetime in the 20-nm-thick Zn_{0.99}Mn_{0.01}Se layers is still much longer than that in the 10-nm-thick layers, but the heating efficiency is similar, see Fig. 4a. At $n_{0}$ the lifetime of holes in the 10/5 nm sample reaches the value of $\tau_{0} \approx 20$ ps, as marked by an arrow and a dashed line in Fig. 4b. This time is sufficient for complete relaxation of the holes reaching a spin temperature below the Mn ion spin temperature. Hence, an upper limit of the characteristic time for spin and energy transfer from the photoexcited holes to the Mn spin system in Zn_{0.99}Mn_{0.01}Se DMS can be estimated by $\tau_{0} \approx 20$ ps.

It is well known that the hole exchange scattering with the localized Mn spin in II–VI DMS is fast, shorter than 1 ps [27]. Hence, the characteristic time measured in our experiments for the Mn heating by photogenerated holes up to 20 ps leads us to conclusion that each hole undergoes multiple scatterings with Mn ions. Such a phenomenon has been found experimentally in (Zn,Mn)Se/(Zn,Be)Se QWs with type-I band alignment, and the responsible mechanism

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**Figure 4** (a) Photo-carrier-density dependence of the reduction of the giant Zeeman shift, $\delta E$, averaged over the time interval $t_{d} = 40$–230 ns. In the inset, $\delta E$ is shown for low carrier concentrations. Note the different half-logarithmic presentations in the main panel and inset. (b) Lifetime $\tau$ of photo-holes in Zn_{0.99}Mn_{0.01}Se layers for each sample in dependence on the photo-carrier density. Solid lines in both panels are guides for the eye.

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**Figure 5** Decay kinetics of PL intensity $I_{D}$ of direct transition for two carrier densities for 10/5 nm (a) and 20/10 nm (b) sample; $T = 1.8$ K and $B = 0$ T. The PL decays are modeled by single-exponential functions shown by the green lines.
has been suggested [17]. The specific feature of this situation is that in external magnetic fields the giant Zeeman splitting of the heavy holes exceeds considerably the hole spin temperature. Accordingly, the initial and final states of the holes participating in the exchange scattering with Mn spins belong to the same spin subband with total angular momentum projection $j_z = +3/2$ along QW growth $z$-axis. For such conditions a single spin flip-flop process between the subbands of a photoexcited heavy-hole and Mn ion is forbidden, while it will be allowed if the mixture between light-hole and heavy-hole states is taken into account.

4 Summary We studied the efficiency of the Mn-spin system heating in dependence on the lifetime of photogenerated holes in Zn$_{0.95}$Mn$_{0.05}$Se layers of (Zn,Mn)Se/(Be,Mn)Te heterostructures with type-II band alignment and different layer thicknesses. The spin and energy transfer from the carriers to Mn ions is mainly governed by photo-holes, and photo-generated electrons are of minor importance. The efficiency of Mn heating depends on the hole lifetime in the (Zn,Mn)Se layer and, particularly, their multiple spin-flip scatterings with localized spins of Mn ions. We demonstrated that the lifetime of the photo-holes, limited by their relaxation into (Be,Mn)Te layers, is sensitive to the excitation power and layer thickness. The observed photogenerated heating of the Mn spin system during $\tau_0 \approx 20$ ps allowed us to define $\tau_0$ as the characteristic time for the direct spin and energy transfer from the photoexcited holes to the Mn spin system in DMS with 1% Mn concentration.

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