phys. stat. sol. (b) **241**, No. 3, 568–571 (2004) / **DOI** 10.1002/pssb.200304303

Spin–lattice relaxation in diluted magnetic (Cd,Mn)Se quantum dots

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Received 25 September 2003, accepted 6 January 2004
Published online 19 February 2004

PACS 72.25.Rb, 73.21.Fg, 73.21.La, 75.50.Pp

Spin–lattice relaxation (SLR) is studied in quantum well and quantum dot structures by photo-carrier induced spin heating. For the SLR time constants no qualitative difference is found between quantum well and quantum dot structures. There is no hint for a specific zero-dimensional behaviour.

1 Introduction Diluted magnetic II–VI quantum dot (DMQD) structures have attracted much interest [1–4], as they have the potential for controlling individual exciton spins on a nm-length scale via the sp-d exchange interaction between interior QD states and localized moments of magnetic ions. Besides this interaction, the coupling of the magnetic ion spins to the phonon system is of fundamental interest. It determines the time scale on which the excited spin system relaxes towards equilibrium. So far, studies on the spin–lattice relaxation (SLR) on diluted magnetic hetero-structures have been restricted to quantum wells (QWs). Recently, a first report on SLR in DMQDs has been published, where a distinct modification in the case of zero-dimensional confinement has been found [5]. In this paper, we perform a SLR measurement on a (Cd,Mn)Se DMQD sample with a Mn content $x = 0.07$. A new technique based on photo-carrier induced spin heating is used providing an improved time resolution. For comparison, the SLR time constants have been measured on (Zn,Cd,Mn)Se QWs of larger thickness.

2 Experimental The DMQD sample has been grown by molecular beam epitaxy on a ZnSe buffer layer on top of a GaAs substrate, applying a thermally activated surface reorganization of a three monolayer $Cd_{0.93}Mn_{0.07}Se$ film [3]. This technique, well established for non-magnetic CdSe QDs, yields a distinct Stranski-Krastanov morphology of three-dimensional islands with a core of pure CdSe [6] placed on a wetting-like layer. Ultra-high vacuum atomic force microscopy proves that the thermally activated reorganization of the DMQD structures results in a monomodally distributed dot ensemble with an average height of 1.2 nm and densities of about 600 μm^{-2}. Finally, the structure is capped by a 85 nm ZnSe layer. Magneto-photoluminescence studies have revealed a giant Zeeman effect with a g-factor of 350 at low magnetic fields ($B < 1$ T) as expected for such a Mn content [7]. The QWs studied have a similar design. Here, the smaller Cd ($y < 0.2$) and Mn ($x < 0.07$) content result in a pseudomorphic growth of much thicker layers. An exact QW thickness is achieved by a RHEED controlled growth.

To observe the relaxation of the Mn spin gas temperature T_s down to the lattice temperature, the spin system is usually heated by non-equilibrium phonons from an electrical or optical generated heat pulse in a metal film. In this study, we take advantage of the fact that even the excitation of photoluminescence can yield a remarkable heaing of T_s, as reported for CdTe [8] and ZnSe [9] QW structures. The scheme of the measurement is sketched in Fig. 1. Using a high-speed acusto-optical modulator with a rise/fall

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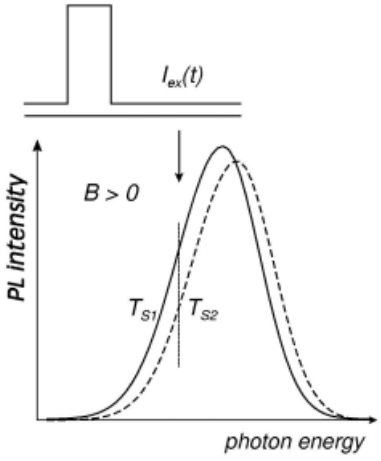

Fig. 1 Scheme of SLR measurements ($T_{s1} < T_{s2}$).

time of 5 ns, the excitation intensity is modulated yielding a pulse with variable pulse width and repetition rate superimposed on an about 10% background excitation intensity.

The detection of the spin gas temperature relies on the temperature dependent Zeeman splitting. At finite magnetic field B, a heating from T_{s1} to T_{s2} results in a reduced intensity at the low-energy side of the PL band. This effect is enhanced in the present structures since an increased T_s reduces also the quantum yield [7]. The temporal shape of the PL is detected by a time-correlated photon counting unit. To calibrate the photo-carrier induced spin heating, stationary PL is measured in a first step in dependence on B for various excitation intensities. The excitation photon energy is chosen in resonance to the barrier exciton state with an excess energy of about 0.37 eV.

3 Results and Discussion Figure 2 displays the shift of the σ^+ PL band versus B for different excitation intensities I_{exc}. Clearly seen is the reduced Zeeman splitting with growing I_{exc}, reflecting the increasing spin gas temperature. To derive the respective values of T_s, the experimental data are fitted by the expected shift of the DMQD exciton groundstate energy with exciton spin +1, i. e. with an electron and hole spin projection of -1/2 and 3/2, respectively, in mean-field approximation [10]

$$\Delta E = \frac{1}{2} N_0 (p_e \alpha_e - p_h \beta_h)\, x\, r(x)\, B_{5/2}\left[\frac{g_e \mu_B B}{k_B (T_0 + T_s)}\right] . \tag{1}$$

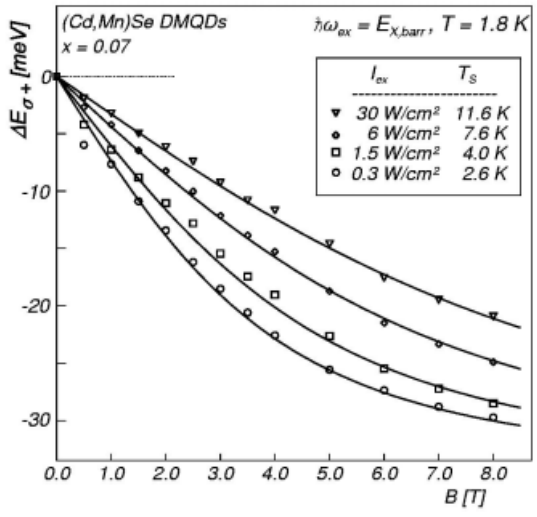

Fig. 2 Energy shift of the σ^+ PL band vs. B for different excitation intensities. The lines represent a fit of the data according to (1). The derived spin gas temperatures are given in the insert.

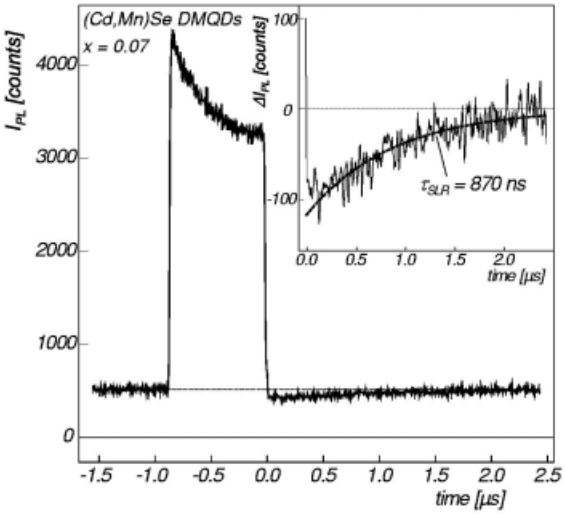

Fig. 3 Temporal shape of the DMQD PL under pulsed excitation as depicted in Fig. 1. Insert: Enlarged view for $t > 0$. The dashed line represents a single exponential fit of the PL intensity change ΔI_{PL}.

Here, the average spin of a Mn ion is described by the $B_{5/2}$-Brillouin function with the Mn electron g-factor g_e and the Bohr magneton μ_B. $N_0\alpha_e$ and $N_0\beta_h$ are the sp-d exchange constants of electron and hole, repectively, in (Cd,Mn)Se [10]. The parameter T_0 and $r(x)$ account for the anti-ferromagnetic pairing of the Mn ions. We have chosen $r(x) = (1–x)^{12}/3 + 2(1–x)^8/3$, which corresponds to the nearest neighbor coordination in the cation fcc sublattice for three monolayers and describes the present DMQD structures quite well [7]. $T_0 = 4.4$ K is derived from the Zeeman splitting at low excitation densities with reduced excess energy [7].

In (1), the finite probabilities to find the carriers in the semimagnetic regions of the dot and wetting layer are explicitely taken into account. From a finite element calculation based on the known morphologic data, we derived $p_e = 0.63$ and $p_h = 0.56$ [7]. The result of the fit given in the inset of Fig.3 proves an efficient heating of the Mn spin system via photo-excited carriers in the zero-dimensional structure. An increase of T_s of about 10 K is achieved at $I_{exc} = 30$ W/cm². We emphasize that the findings are qualitatively very similar to data observed on ZnSe QWs [9].

In Fig. 3, the temporal shape of the PL on the low-energy side (marked by an arrow in Fig. 1) is depicted for pulsed excitation. The progressive PL decrease at the plateau of the excitation pulse ($–800$ ns $< t < 0$ ns) reflects the Mn spin heating associated with a high-energy shift of the PL band. After switching back to the background excitation intensity ($t = 0$), the PL recovers to the equilibrium value. In the insert, an enlarged view is given for this range. From an single-exponential fit of the PL change, a SLR time constant of 870 ns is derived. For higher excitation intensities, a non-exponential behavior is observed both in the heating and cooling period, which needs further investigations.

Table 1 SLR time constant τ_{SLR} of diluted magnetic quantum wells and dots with different Mn content x.

Sample	x	τ_{SLR} [µs]
(Zn,Cd,Mn)Se QW	0.02	5.5
(Zn,Cd,Mn)Se QW	0.07	1.2
(Cd,Mn)Se DMQD	0.07	0.9

In Table 1, this time constant is compared with those measured by the same technique on (Zn,Cd,Mn)Se QWs. The QW data reflect the expected tendency that the SLR rate grows exponentially with the Mn content, caused by the contribution of Mn pairs and higher complexes in the coupling of the Mn spins to the lattice [11]. We emphazise that the values observed on the quarternary (Zn,Cd,Mn)Se QWs match well with data reported for (Zn,Mn)Se QWs [12]. The DMQD time constant deviates only marginally from the QW data with the same nominal x. No hint on a slow non-exponential component as reported in [5] is observed. We conclude that in the studied DMQD sample the SLR takes place locally in a similar way as in QW and bulk samples.

Acknowledgements This work has been supported by the Deutsche Forschungsgemeinschaft within the point of interest SP1133.

References

[1] C. S. Kim, M. Kim, S. Lee, J. Kossut, J. K. Furdyna, and M. Dobrowolska, J. Cryst. Growth **214/215**, 395 (2000).
L. V. Titova, J. K. Furdyna, M. Dobrowolska, S. Lee, T. Topuria, P. Moeck, and N. D. Browning, Appl. Phys. Lett. **80**, 1237 (2002).
[2] Y. Terai, S. Kuroda, and K. Takita, Appl. Phys. Lett. **76**, 2400 (2000).
[3] P. R. Kratzert, J. Puls, M. Rabe, and F. Henneberger, Appl. Phys. Lett. **79**, 2814 (2001).
[4] N. Takahashi, K. Takabayashi, I. Souma, J. Shen, and Y. Oka, J. Appl. Phys. **87**, 6469 (2000).
[5] A. V. Scherbakov, D. R. Yakovlev, A. V. Akimov, W. Ossau, L. W. Molenkamp, T. Wojtowicz, G. Karczewski, J. Kossut, J. Cibert, S. Tatarenko, Y. Oka, and I. Souma, phys. stat. sol. (b) **229**, 723 (2002).
[6] D. Litvinov, A. Rosenauer, D. Gerthsen, P. Kratzert, M. Rabe, and F. Henneberger, Appl. Phys. Lett. **81**, 640 (2002).

[7] A. Hundt, J. Puls, and F. Henneberger, Phys. Rev. B, Rapid Commun., submitted.
[8] M. G. Tyazhlov, V. D. Kulakovskii, A. I. Filin, D. R. Yakovlev, A. Waag, and G. Landwehr, Phys. Rev. B **59**, 2050 (1999).
A. V. Koudinov, Yu. G. Kusraev, and I. G. Aksyanov, Phys. Rev. B **68**, 085315 (2003).
[9] D. Keller, D. R. Yakovlev, B. König, W. Ossau, Th. Gruber, A. Waag, L. W. Molenkamp, and A. V. Scherbakov, Phys. Rev. B **65**, 035313 (2001).
[10] O. Goede and W. Heimbrodt, phys. stat. sol. (b) **146**, 11 (1988).
[11] D. Scalbert, phys. stat. sol. (b) **193**, 189 (1996).
[12] D. R. Yakovlev, M. Kneip, A. A. Maksimov, I. I. Tartakovskii, M. Bayer, D. Keller, W. Ossau, L. W. Molenkamp, A. V. Scherbakov, A. V. Akimov, and A. Waag, phys. stat. sol. (c) **1**, No. 4 (2004), this conference.