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Spin-lattice relaxation in semimagnetic CdMnTe/CdMgTe quantum wells

A. V. Scherbakov,[1] A. V. Akimov,[1] D. R. Yakovlev,[1,2] W. Ossau,[2] G. Landwehr,[2] T. Wojtowicz,[3] G. Karczewski,[3] and J. Kossut[3]

[1]*A.F. Ioffe Physical-Technical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia*

[2]*Physikalisches Institut der Universität Würzburg, 97074 Würzburg, Germany*

[3]*Institute of Physics, Polish Academy of Sciences, 02-668 Warsaw, Poland*

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The spin-lattice relaxation dynamics of magnetic ions has been studied in semimagnetic semiconductor $Cd_{1-x}Mn_xTe/Cd_{0.88}Mg_{0.12}Te$ quantum wells ($x \leqslant 0.032$), by means of a method based on optical detection of injected nonequilibrium phonons. The relaxation rate is an increasing function of Mn content, external magnetic field and lattice temperature. The energy of phonons active in the spin-lattice relaxation process is found to increase with magnetic field, which can be explained in terms of phonon-induced spin-flip transitions in clusters consisting of the next-nearest-neighbor magnetic ions.

Static magnetic properties of diluted magnetic semiconductors (DMS's), e.g., the magnetic susceptibility, the occurrence of magnetization steps, the specific heat have been widely investigated in the past years and are relatively well understood.[1] However, *dynamical* magnetic properties of DMS's caused by spin-spin and spin-lattice interactions, have been adequately investigated. This situation is very undesirable in view of a growing interest in DMS materials as promising components of future spin-electronic devices, which exploit the spin states of free carriers for digital operations.[2,3] We have reported recently that in $Cd_{0.99}Mn_{0.01}Te$-based quantum wells (QW's) with a slow spin-lattice relaxation (SLR) rate the magnetic ion system can be substantially overheated by means of interaction with hot photocarriers, which is enhanced by the presence of a two-dimensional electron gas.[4] Effective mechanisms providing SLR are important to keep the system of magnetic ions in thermodynamical equilibrium with the lattice.

The SLR in Mn-based DMS's has been studied experimentally by several groups.[5–9] These experiments were performed on bulk DMS samples with Mn content in the range $x=0.002-0.05$, at lattice temperatures T_0 varying from 1.5 to 30 K and in magnetic fields up to 25 T.[8] The results of those experiments are briefly summarized here: (i) the SLR rate τ^{-1} increases weakly in magnetic fields B below 12 T (Refs. 5, 6, and 9) and increases rapidly ($\tau^{-1} \propto B^5$) for $B > 17$ T;[8] (ii) τ^{-1} is an increasing function of the lattice temperature T_0; (iii) the phonon bottleneck effect is observed at low T_0;[5–7] (iv) τ^{-1} is an increasing function of the Mn ion concentration; (v) peculiarities in τ^{-1} vs B dependence were observed at $B \approx 12$ and 19 T,[7,8] i.e., at fields where the singlet and the triplet state of a nearest-neighbor pair of antiferromagnetically interacting Mn ions cross. All authors have noted the importance of antiferromagnetic clusters for the SLR. Also the role of defects and impurities cannot be excluded.[10] Scalbert suggested the particular SLR mechanism via an Orbach process in a cluster of three nearest-neighbor Mn ions.[11] However, this mechanism alone does not allow a full description of experimental results at low temperatures ($T_0 < 5$ K).

At present, there are no experimental data for SLR rates measured at $T_0 < 2$ K and at magnetic fields $B < 10$ T that would be free of the phonon bottleneck effect. This effect does not allow to obtain experimental determination of the rate of elementary spin-phonon transitions. A phonon bottleneck effect occurs when the heat capacity of the magnetic system is higher than the heat capacity of the phonon reservoir.[12] Then the cooling (heating) rate of the spin system is governed not only by the SLR process, but also by the phonon dynamics. However, the phonon bottleneck effect can be ruled out in those QW structures where the phonons in massive substrates and thick nonmagnetic barriers [e.g., in (Cd, Mn)Te/(Cd, Mg)Te QW's] are hardly affected by the phonon emission/absorption processes in the spin-containing thin DMS layers. So far no studies of the SLR in QW structures were performed.

An experimental method is exploited in this paper for the investigation of the SLR dynamics in (Cd, Mn)Te/(Cd, Mg)Te QW's. It is a combination of nonequilibrium phonon- and exciton-luminescence techniques.[13,14] The optical detection of the spin-system dynamics allows us to overcome problems of low sensitivity, which are characteristic for methods based on direct measurements of the magnetization. These methods are not suitable for studying structures with a single QW with a thickness of a few monolayers. We show that the phonon energy, which is relevant in SLR processes in DMS's increases with a magnetic field. This important result allows a better understanding of the SLR mechanism in a DMS, which involves antiferromagnetic clusters.

The QW structures studied were grown by molecular beam epitaxy on 0.4-mm-thick semi-insulating GaAs substrates with a 1 μm $Cd_{0.88}Mg_{0.12}Te$ buffer layer. The structure, on which the results reported here are obtained, contains three 23-nm-thick $Cd_{1-x}Mn_xTe$ QW's with $x=0.016$, 0.025, and 0.032. (The Mn content has been evaluated from the giant Zeeman splitting of excitons measured in reflectivity spectra.) The QW's were separated by 50-nm-thick nonmagnetic $Cd_{0.88}Mg_{0.12}Te$ barriers. Nonequilibrium phonons were generated by the well-known heat pulse technique,[15] a schematic of the experimental setup is shown in Fig. 1. The phonon generator h (a 10-nm-thick constant film) with an

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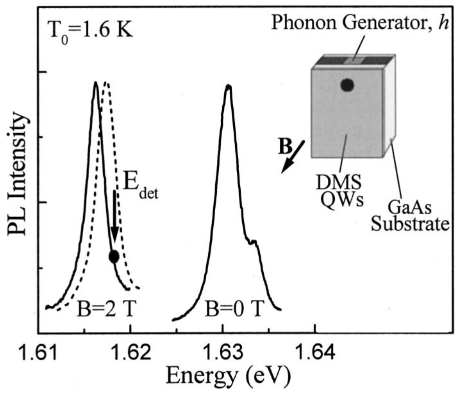

FIG. 1. Exciton luminescence spectra of $Cd_{0.975}Mn_{0.025}Te/Cd_{0.88}Mg_{0.12}Te$ QW's measured in the absence (solid lines) and in the presence (dashed line) of nonequilibrium phonons. E_{det} shows the energy where the phonon-induced PL intensity signal $\Delta I(t)$ was detected [see Fig. 2(b)]. The inset shows the experimental scheme. A black spot is an area where the SLR time and T_S are measured (see details in Ref. 14).

area $0.5\times 0.25\,\mathrm{mm}^2$ was evaporated on the narrow edge of the GaAs substrate and was heated by current pulses of duration $\Delta t = 0.1\,\mu$s at a repetition rate of 5 kHz. The pulse power density P was varied from 30 up to 180 W/mm^2. The phonons generated in h during the current pulse have a Planck distribution, characterized by a heater temperature $T_h \propto P^{1/4}$. The value of T_h can be calculated from the acoustic mismatch theory.[16] The result is a broad spectrum of nonequilibrium phonons with characteristic energies of several meV (frequency $\sim 10^{11}-10^{12}$ Hz) which is injected into the sample. These phonons propagate in the GaAs substrate, reach the DMS QW and heat the spin system of the magnetic ions. The duration of the phonon excitation at the detection point is considerably longer than the duration of the current pulse, it is about 2–3 μs.[14] It is assumed that due to fast ($\sim 10^{-8}$ s) spin-spin interaction processes in DMS the Mn spin temperature T_S is established. We measure T_S in a small area (0.2 mm in diameter—as determined by the spatial extension of the focused laser beam) located 0.3 mm away from the center of the phonon generator. T_S is determined from the value of the giant Zeeman splitting of excitonic states, which causes a strong energy shift of exciton line in photoluminescence (PL) spectra (Fig. 1). PL spectra were dispersed with a 1 m spectrometer and detected by a fast photomultiplier. Analysis with the time resolution up to 10 ns was achieved by means of a special interface board and a computer. The time-resolved detection of $T_S(t)$ is based on monitoring the dynamical shift of the exciton PL line (for details see Refs. 13 and 14). Low photoexcitation power (<0.1 W/cm^2) was chosen to avoid additional heating of the Mn system.[4] The sample was mounted inside a superconducting magnet in the Faraday configuration and was immersed in liquid helium.

Figure 2 shows the normalized phonon-induced signal $\Delta I(t) = I(t) - I_0$ (here I_0 is the PL intensity without phonon injection) measured at $T_0 = 1.6$ K at different magnetic fields. We note here that in the linear regime $\Delta I(t)$ is proportional to the variation of the Mn spin temperature $\Delta T_S(t)$ (for details see Ref. 14). In this paper we will discuss the data of Fig. 2 assuming that they reflect dynamical changes of the

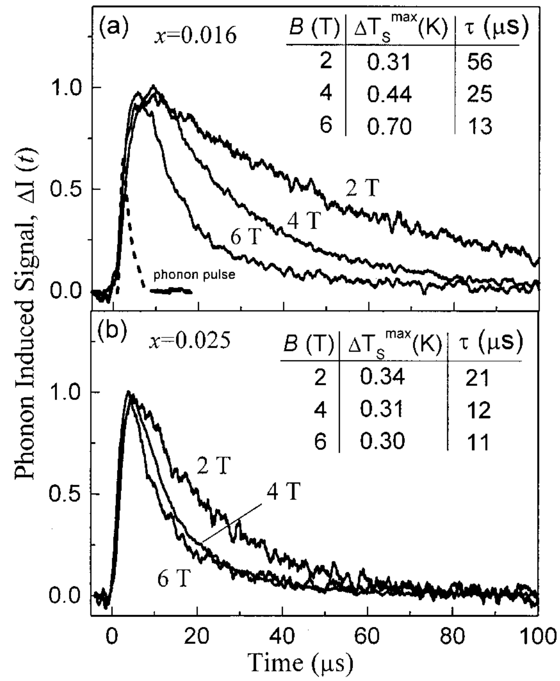

FIG. 2. Time evolution of the phonon-induced variation of the PL intensity (corresponding to variation of the Mn spin temperature) detected on the high energy side of the PL line (see Fig. 1) for different magnetic fields and two concentrations of Mn ions. Signals are normalized on their maximum intensities. Temperatures of phonon generator $T_h = 25$ K (a) and 17 K (b). The dashed curve shows the time evolution of nonequilibrium phonons in the detection area (the vertical scale is arbitrary). $T_0 = 1.6$ K.

spin temperature. The leading edges of $\Delta T_S(t)$ have a width of about 2 μs which is about the duration of the phonon pulse. The decay times τ, determined from the exponential fit of $\Delta T_S(t)$, are listed in the tables in Fig. 2 together with the maximal temperature increase ΔT_S^{max} of the Mn system. Obviously, the time τ is the cooling time of the Mn spin system and in our case, where the phonon bottleneck is absent, it is actually SLR time of the Mn ions at the bath temperature T_0.

The SLR times exhibit strong dependencies on x, B, and T_0. In Fig. 3(a) the SLR rate τ^{-1} dependence on a magnetic field is shown for QW's with different Mn content. The strongest increase of the SLR rate of five times with B is observed in the QW with the lowest Mn content of $x = 0.016$. Elevation of the lattice temperature from 1.6 K to 4.2 K leads to an increase of τ^{-1} but does not affect the observed magnetic field dependence. In Fig. 3(b), τ^{-1} is plotted as a function of the Mn content for $B = 2$ T. The SLR rate increases drastically (by an order of magnitude from $10^4\,\mathrm{s}^{-1}$ to $10^5\,\mathrm{s}^{-1}$) with a rather small variation of the Mn content from 0.01 up to 0.032. Such a behavior, reported previously for bulk materials,[5–9] provides evidence for the dominating role of the Mn spin clusters in the SLR process. We would like to stress that our measurements of SLR rates are free of the phonon bottleneck effect. This is likely to be a reason for the different values of τ^{-1} measured in the earlier studies with bulk DMS's.[5–9]

Let us now analyze the amplitudes of phonon-induced signals, which reflect absolute values of $\Delta T_S(t)$. It is important that in our experiments the phonon spectrum at the detection point is not in equilibrium, i.e., can not be described

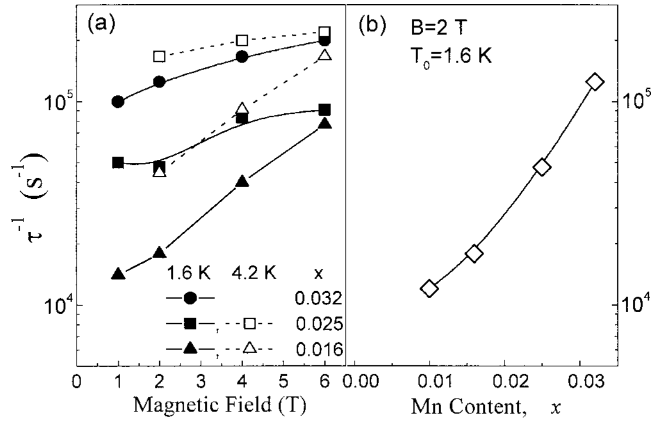

FIG. 3. (a) A dependence of SLR rates on a magnetic field for $Cd_{1-x}Mn_xTe/Cd_{0.88}Mg_{0.12}Te$ QW's with different x at $T_0=1.6$ K (closed symbols) and 4.2 K (open symbols). (b) SLR rate as a function of Mn content. Lines are guides for the eye. Error bars are smaller than the size of the symbols. $T_h=17$ K.

by the Planck distribution with a unique temperature. However, it can be described by an effective temperature of phonons $T_{\rm eff}(\omega)$, which depends on the phonon energy $\hbar\omega$:

$$\frac{1}{\exp(\hbar\omega/k_BT_{\rm eff})-1}=\frac{\alpha(\omega)}{\exp(\hbar\omega/k_BT_h)-1}+\frac{1}{\exp(\hbar\omega/k_BT_0)-1}. \qquad (1)$$

The coefficient $\alpha(\omega)$ is governed by an energy-dependent scattering of phonons in the GaAs substrate and at interfaces, it depends also on the distance from h to the detection area.[17] In our experiments we estimate a maximum value of $\alpha(\omega)=0.05-0.1$. In the range of the phonon energies ($\hbar\omega=0.1-1$ meV) of interest to us, $\alpha(\omega)$ is known to be a decreasing function of $\hbar\omega$ due to a strong energy dependence of the phonon reflection at the crystal/liquid helium boundary.[18] It follows from Eq. (1) that $T_{\rm eff}(\omega)$ *decreases with an increase of* $\hbar\omega$.

Now we turn to the analysis of phonon energies active in the SLR process in various magnetic fields. From the time evolution of $\Delta T_S(t)$ in the spin system, presented as $\Delta I(t)$ in Fig. 2, one can obtain the time evolution of $T_{\rm eff}(t)$ in the phonon system using the convolution equation:

$$T_S(t)=\frac{1}{\tau}\int_0^t T_{\rm eff}(t')\exp\left(-\frac{t'-t}{\tau'}\right)dt', \qquad (2)$$

where $T_S(t)=T_0+\Delta T_S(t)$. The result of deconvolution of Eq. (2) gives us $\Delta T_{\rm eff}(t)=T_{\rm eff}(t)-T_0$, which is shown in Fig. 2(a) by a dashed line. We have found that the shape of $\Delta T_{\rm eff}(t)$ does not depend on B and x, which confirms that the phonon bottleneck is absent and the spin system does not influence the phonon time evolution. We evaluated the mean effective temperature during the phonon pulse $\overline{T}_{\rm eff}$. Its magnetic field dependencies for QW's with $x=0.025$ and 0.032 are plotted in Fig. 4. $\overline{T}_{\rm eff}$ decreases in stronger magnetic fields. Keeping in mind that $T_{\rm eff}$ is a decreasing function of $\hbar\omega$ we conclude that the *energy of phonons, which are active in the heating of spin system, increases with the increase of* B. We note here that the same result was obtained earlier for DMS QW's with $x=0.07$, where SLR dynamics were faster than that of the phonons.[14]

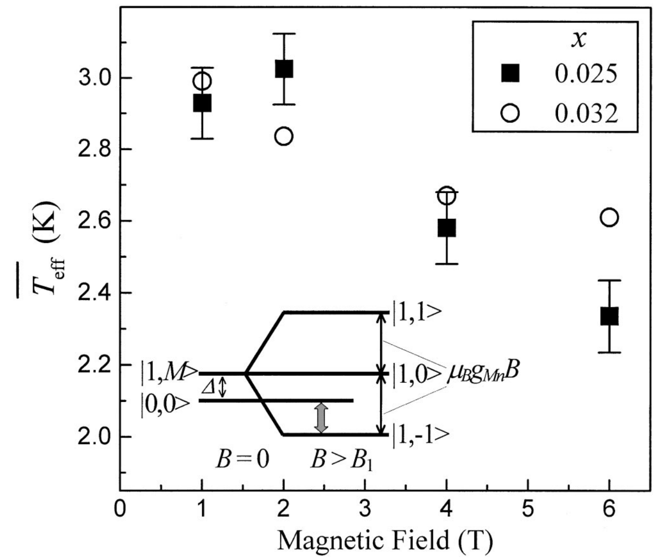

FIG. 4. A magnetic field dependence of $\overline{T}_{\rm eff}$ for nonequilibrium phonons which are active in the heating of the Mn spin system. ($T_0=1.6$ K, $T_h=17$ K). A scheme of energy levels and spin-phonon transitions in a pair of magnetic ions are shown in inset.

Starting the discussion we first rule out Raman processes as the principal SLR mechanism. Raman processes have very strong temperature dependences of τ^{-1} ($\tau^{-1}\sim T_0^n$, where n is between five and nine, depending on the type of magnetic ion).[12] This is definitely not the case of the studied samples at $T_0<4.2$ K (see Fig. 3). We suggest that the SLR mechanism is due to direct spin-phonon transitions. Our main aim is to find the mechanism, which allows the effective spin-phonon transitions accompanied by the changes of the spin polarization.

Spin-phonon transitions in isolated Mn^{2+} ions ($S=5/2$, $L=0$) cannot account for the measured values of τ, due to $L=0$ electronic states of Mn ions interacting very weakly with the lattice.[12] Thus, the SLR process requires the participation of Mn-ion clusters. In such clusters a system of spin levels is formed due to spin-spin superexchange interaction. The spin-phonon transitions $\Delta S=\pm 1$; $\Delta M=0,\pm 1$ between these levels are allowed and governed by the Dzyaloshinski-Moriya mechanism.[7,11,19] For example, the singlet ($S=0$) to triplet ($S=1$) transitions between the lowest spin levels in a pair of Mn ions are shown in the inset of Fig. 4. At the magnetic field $B>B_1$ ($B_1=\Delta/\mu_B g_{\rm Mn}$ is the field where the $|0,0\rangle$ and $|1,-1\rangle$ states cross each other) the absorption of resonant phonons by $|1,-1\rangle\rightarrow|0,0\rangle$ transition causes a heating of spin system and a decrease of the magnetization. The reverse $|0,0\rangle\rightarrow|1,-1\rangle$ transition is accompanied by the emission of a resonant phonon and increases the magnetization. It is clearly seen from Fig. 4 that at $B>B_1$ the phonon energy $\hbar\omega=\mu_B g_{\rm Mn}B$ being resonant with the $|0,0\rangle\leftrightarrow|1,-1\rangle$ splitting increases with B. Such behavior is consistent with the experimental fact that the energy of a phonon active in the SLR process increases with B.

The case of nearest-neighbor pairs and triads of Mn ions is, however, not applicable to our experiments because of a too high value of $B_1 > 10$ T.[1,7,8,19] However clusters consisting of next-nearest-neighbor Mn ions have essentially a lower value of B_1 and may contribute to the present experimental observations (especially if we take into account the small values of Mn content in the investigated samples). For example, for next-nearest-neighbor pairs of Mn ions $B_1 \approx 2$ T (Ref. 1) and in the field range $B = 2-7$ T used in our experiments the requirement $B > B_1$ is met.

Obviously, in DMS materials a variety of clusters with different values of B_1 (Ref. 20) can exist, however not all of them will be active in SLR process. Further theoretical studies are needed to understand the role of different clusters in detail. Experiments with monochromatic phonons would be very useful for a comprehensive understanding of SLR mechanisms in DMS's.

In conclusion, we have shown, that it is possible to obtain information on the spin-lattice relaxation dynamics in heterostructures of diluted magnetic semiconductors by employing nonequilibrium phonons. The method, due to optical detection, is highly sensitive, so that it has been possible to investigate thin layers with a thickness of a few monolayers. It should be possible to apply the method to single quantum dots. The measured values of the spin-lattice relaxation rate in quantum wells as a function of magnetic field, temperature and Mn content do not suffer from the phonon bottleneck effect. We hope that our work will stimulate the development of theoretical models, necessary for the quantitative explanation of spin-lattice relaxation.

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