Light-induced heating effects in semimagnetic quantum wells

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A simple explanation of the low-temperature light-induced heating effects in semimagnetic quantum wells is suggested. It is shown that one ought not to disregard the heating of the crystal lattice by the optical excitation under typical conditions of the magneto-optic experiments.

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The quantum-well structures with semimagnetic layers are of great interest since they can lead to the production of highly spin-polarized carrier populations, for which several types of applications have been proposed. These giant values of carrier spin polarizations, as well as of the excitonic Zeeman (spin) splittings, are due to the effect of the ''exchange field'' of magnetic ions (which are spin aligned with the magnetic field) on the carrier/excitonic spins. In effect, both the excitonic spin polarization and the value of the excitonic spin splitting monitor the degree of the spin alignment of magnetic ions in an external magnetic field.

Recently, König *et al.*[1] reported on the effects of light-induced heating in a quantum well (QW) made of a diluted magnetic semiconductor (DMS), containing a 2D electron gas (2DEG), and placed into an external magnetic field. The main observation of the paper is a significant decrease of the Zeeman splitting of excitonic states with increase of power of the pumping laser beam. The change of the Zeeman splitting was observed in both photoluminescence and reflection spectra. In view of aforementioned considerations, the natural explanation of the effect is a light-induced heating of the system of magnetic-ion spins, which leads to their thermal disorder, leading, in turn, to the decrease of the ''exchange field.''

The authors of Ref. 1 have put forward a specific mechanism of light-induced heating, in which a 2DEG existing in the QW is considered as a mediator of the energy transfer from photogenerated carriers to manganese spins. This mechanism was supposed to be the main one, while the usual heating of the phonon system, i.e., of the crystal lattice, was neglected.

Meanwhile, in the magneto-optic studies of DMS's, the effects of heating by light have been known for many years.[2–6] In particular, when the magnetic-field-induced circular polarization of luminescence is being measured, the polarization degree depends on optical pump density (PD), which is routinely observed in QW's nominally containing no extra electrons, and also in undoped bulk crystals as well. The fact worth noting is that this effect is usually rather strong (the polarization varies by several times) and occurs roughly in the same PD domain as the effect reported in Ref. 1. Faced with this effect again and again, we recently attempted to clarify its origin and performed several experiments for this purpose. The results convinced us that in our samples, the phonon system is responsible for the heating of manganese ions, i.e., the usual thermal action of light takes place.

The present paper has the following structure. First, the samples used by us are described. Then, we present the experimental results which bridge the gap between the observations of Ref. 1 and the effect which we observed in our undoped samples. Further, when the analogy between the effects is established, we show that the explanation of our results based on the model of Ref. 1 runs into difficulties. Finally, the alternative interpretation is discussed.

I. SAMPLES

We used three samples, each containing a set of isolated QW's. Samples A1 and A4 were grown according to a common design, the only difference between them being the different manganese concentrations (1 and 4 %, respectively). Namely, the semi-insulating GaAs substrates were covered with 5 μm $Cd_{0.88}Mg_{0.12}Te$ buffers followed by 0.5 μm $Cd_{0.87-x}Mn_xMg_{0.13}Te$ and further, by sandwiches of 100, 60, and 40 Å $Cd_{1-x}Mn_xTe$ QW's alternating with 300 Å $Cd_{0.87-x}Mn_xMg_{0.13}Te$ barriers. Sample B7 was grown on $CdZn_{0.036}Te$, with $Cd_{0.93}Mn_{0.07}Te$ QW's of 300, 80, 45, 18, and 9 Å being separated from each other with 500 Å $Cd_{0.64}Mn_{0.07}Mg_{0.29}Te$ barriers.

II. HEATING DETECTED BY POLARIZATION

At low concentrations of magnetic (manganese) ions the DMS's behave as paramagnets, i.e., the manganese spins tend to align with the applied magnetic field. The spin polarization of electrons and holes forming excitons occurs in the ''exchange field'' of spin-aligned manganese ions, and it is this spin polarization which manifests itself in circular polarization of the excitonic luminescence.

Figure 1(a) shows a typical manifestation of the heating effect in the polarized photoluminescence (PL) of the DMS QW's. When the power of the laser beam which excites the luminescence from a QW is increased, the polarization degree decreases—by typically 2–3 times in the ordinary conditions of the PL experiment. Figure 1(b) indicates that the pattern of polarization uprise in the field remains unchanged for a wide range of pump power values. Since the polarization degree taken on the linear part of this pattern is known, in samples with low manganese content, to depend strongly on temperature ($\propto T^{-1}$, Ref. 7), the decrease in polarization is naturally ascribed to an increase of temperature and an associated decrease of magnetization according to the Curie-Weiss law. In effect, the polarization degree is a thermometer

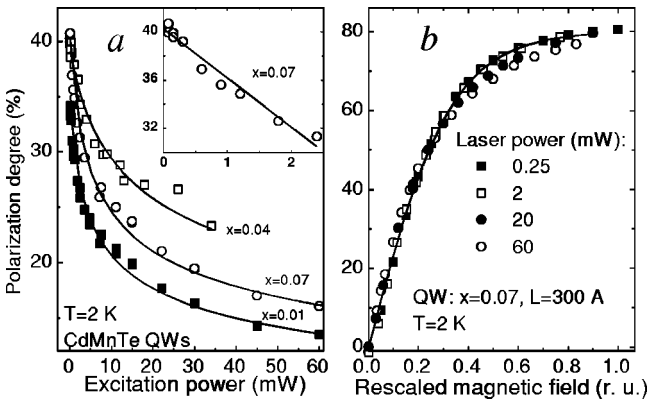

FIG. 1. (a) Magnetic-field-induced PL polarization degree versus optical pump power for samples A1 ($x=0.01$, QW 60 Å, magnetic field 260 Oe), A4 ($x=0.04$, 60 Å, 260 Oe), and B7($x=0.07$, 300 Å, 170 Oe); the inset shows the low-pump region of the latter dependence. We estimate 1 mW of laser power as 0.2 W/cm^2. (b) Scaling of the magnetic field dependences of the PL polarization, measured at different values of pump power. For example, to bring the 0.25 mW curve together with the 60 mW one, we had to stretch it triply along the field scale. For true values of field for each laser power, compare with panel (a). The line shows a hyperbolic tangent fit.

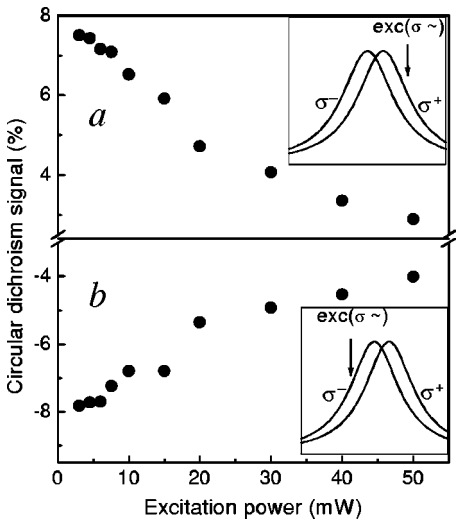

FIG. 2. Magnetic-field-induced circular dichroism in the PLE signal from sample B7, QW 300 Å, vs pump power. The excitation was into (a) the long- and (b) the short-wavelength side of the 1hh exciton. The luminescence intensity was recorded lower in energy.

measuring the temperature of the ensamble of Mn spins.

A change of focusing of the laser on to the crystal immediately results in the shift of the dependences similar to the ones in Fig. 1(a) to another pump range. This means that the heating is governed by PD rather than by full laser power, i.e., the heating envelopes not the crystal as a whole, but (at least predominantly) the irradiated zone. One can ask, ''is the overheating homogeneous within the laser spot or is the system of manganese ions heated locally in some neighborhood of the recombination centers?''

The answer follows from the result displayed in Fig. 2. In this experiment, the PL was excited by a tunable laser close to the resonance of the excitonic transition, alternately (at a high frequency) with left- and right-hand polarized light (σ^+ and σ^-). In fact, here the circular dichroism of excitonic absorption is measured. The recorded signal (bare PL intensity) is proportional to the difference of density-of-states, or absorption coefficients, for the σ^+ and σ^- light. This difference, in turn, is due to the Zeeman splitting of the spectral lines originating from the $+1$ and -1 projections of the angular momentum. The insets in Figs. 2(a), 2(b) help to understand why this signal should have (as it does) opposite signs at the long- and short-wavelength edges of the excitonic line, while in general the behavior of the PLE dichroism is clearly similar to that of PL polarization [Fig. 1(a)]: the signal drops down with PD. However, the essential difference between these two experiments is that the PL polarization is sensitive to localized (radiative) excitonic states, while the PLE dichroism manifests the spin splitting of delocalized (free) excitonic states from which the excitonic absorption band is formed. Since heating alters the spin splitting of delocalized states, one has to conclude that it expands to the whole square of the light spot rather than only to the vicinity of the recombination centers.

As a matter of fact, the two experiments discussed (PL polarization and PLE dichroism) are close analogs of the experiments of Ref. 1 with the splitting of the ''trion'' luminescence line and the splitting of the reflection spectrum, respectively. All our results reported above do not contradict those of Ref. 1; vice versa, the similarity of the experimental manifestations arouses suspicion that in these two cases, the same effect was observed though somewhat different experimental techniques were used.[8]

III. RES CONTRA

The experimental fact which is difficult to explain in terms of Ref. 1 can be briefly formulated as follows: we were able to observe the heating in all of our samples, namely, in A1, A4, and B7, at nearly the same conditions. Our A1 sample differs from the sample studied in Ref. 1 in that it was not intentionally doped with donors in the barrier layer. In Ref. 1, the explanation of the heating involves a 2DEG with concentration 1.2×10^{10} cm^{-2}, which is further suggested to increase with PD up to 3.2×10^{10} cm^{-2}. In our undoped samples, the effect is observed both in cases of above-the-barrier and below-the-barrier excitation. In the latter case the photoassisted inflow of extra electrons into the QW is less probable. An optimistic estimate of the concentration of casual extra electrons in our QWs yields several units of 10^9 cm^{-2}, while the heating effect is just as strong as in Ref. 1, occuring in the same PD domain.

Further, even if one supposes that in our QW's a high concentration of extra electrons exists without intentional doping, it also does not help. The mechanism of heating via the 2DEG is very sensitive to manganese content x. When x is increased, the spin-lattice relaxation time falls dramatically, so that, as can be seen from Fig. 11 of Ref. 1, even for a 2DEG as dense as 10^{11} cm^{-2}, heating can hardly be expected for x greater than 1.5–2%. Contrary to that, we ob-

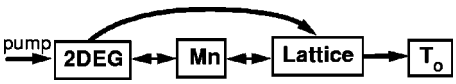

FIG. 3. Scheme of the heat transfer from the 2DEG (and/or photocreated excitons) to the thermostat T_0 (helium bath, cold parts of the sample).

served the effect in samples with higher x.

In our experiments, the heating effect looked very similar for samples A1 and A4, which differed only in manganese concentration [see Fig. 1(a)]. This fact is worth interpreting. As for A4 ($x=4\%$) the manganese reservoir (Mn) is expected to be coupled to the lattice more strongly than to the 2DEG of a reasonable concentration,[1,9] the left two-forked arrow in Fig. 3 is broken and the Mn temperature should be bound to the lattice one. Therefore, even if the 2DEG is present in a noticeable amount, no heating of the Mn would occur, on increase of pumping, if this increase would only result in increase of the temperature offset between the 2DEG and the lattice. Overheating of the lattice compared to the thermostat is required (see Fig. 3). For sample A1 ($x=1\%$), the Mn-lattice coupling is weaker by two orders of magnitude and could become comparable with the Mn to 2DEG coupling,[9] allowing a rise of the Mn temperature in between that of the 2DEG and that of the lattice. However, no significant difference in Mn temperature was observed for A1 and A4 at equal pumping. This can be explained in two ways: either (i) the coupling of Mn to the lattice in A1 is still stronger than to the 2DEG because of too high x or too low 2DEG density or (ii) there is no noticable temperature offset between the 2DEG and the lattice. Whatever the case, the main reason of the observed Mn heating is also the temperature offset between the lattice and the thermostat. In other words, the lattice must be overheated in this (A1) sample too.

IV. HEATING OF SUBSTRATE

In the experiments with photoinduced heating, it is dangerous to fail to bear in mind that the QW itself and its neighborhood absorb only a part of the incident light. At the most accurate (in respect to inflow of electrons from the barrier, etc.) experiments with below-the-barrier excitation, the QW absorbs as little as a few percent of the incident power, while the rest transmits, being absorbed either in the buffer or in the substrate. Naturally, if the PD is increased, it will finally result in heating of the substrate, and via the substrate, the QW region. The only question is ''will the substrate be heated earlier (i.e., at lower PD's) than some other heating effect occurring directly in the QW, or will this latter process come first?''

We have performed the following experiment. We have superimposed two laser beams, the first with higher energy of quanta and low PD (probe beam) and the second, with lower energy of quanta and higher PD (pump beam). The weak probe beam, which did not cause any overheating, was used to excite luminescence from the QW. The polarization of this luminescence was measured by us while the frequency or the power of the pump beam was varied. The

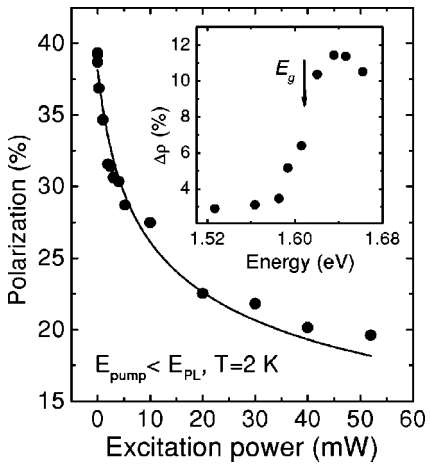

FIG. 4. Polarization of the PL excited by the probe beam vs the pump beam power in sample B7, QW 300 Å; the pump light quantum energy was below the QW states. The inset shows the change in the effect of the pump light on the polarization degree when the pump quantum energy is tuned around the fundamental absorption in the substrate.

results obtained for sample B7 are the most illustrative (Fig. 4). It turns out that a strong heating occurs even when the pump laser frequency is on the long-wavelength side of the PL line, i.e., passing through the QW without noticeable absorption. However, this heating does manifest a clear ''red edge'' which coincides with the fundamental absorption edge of the substrate (see inset in Fig. 4). It is evident that in this case, the substrate heating is a dominating effect. In samples A1 and A4, we also were able to observe heating by light for which both the barriers and the QW's were transparent. However, for these samples we could not attribute all the magnitude of the effect to substrate heating. Most likely, this observation can be explained by the higher heat conductance of more crystallographically perfect GaAs substrates, i.e., by effective evacuation of heat from the front interface of the substrate into the bulk. (We note that the buffer layers in these samples were transparent, contrary to the that in the sample studied in Ref. 1.)

V. OTHER POSSIBLE MECHANISMS

In samples A1 and A4, apart from substrate heating, other heating mechanisms arise when the pump beam is tuned to the short-wavelength side of the exciton in a QW. As it is discussed above, observation of the effect in sample A4 practically excludes heating via a 2DEG. On the other hand, the similarity of effects observed in A1 and A4 can be naturally explained if one supposes that, in both samples, the crystal lattice is locally overheated. So, for these samples the facts also argue in favor of heating mediated by phonons, but now being generated directly in the QW rather than in the substrate.

Figure 5(a) shows a set of heating curves for the 60 Å QW in structure A1, measured at below-the-barrier excitation. The laser frequencies (the points in the PL excitation spectrum) were chosen in such a way that the PL intensity (i.e., concentration of photocreated carriers) stayed nearly

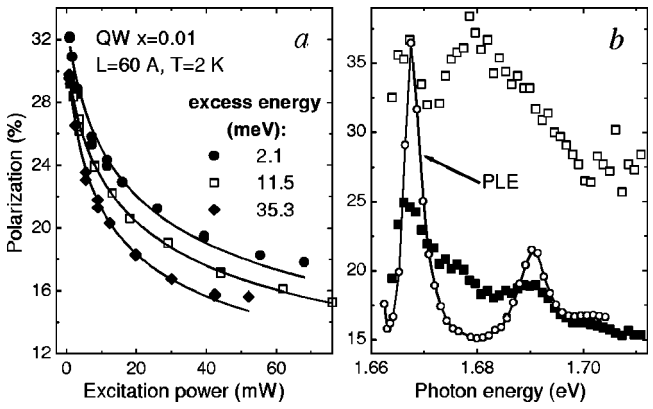

FIG. 5. (a) Polarization vs pump power, as in Fig. 1(a), at different excess energies of exciting quanta. (b) PL polarization vs energy excess, measured at fixed lower (open squares) and higher (filled squares) PL intensity. The PLE spectrum is also shown.

unchanged. In Fig. 5(b) we plotted the PLE spectrum of the same QW together with the polarization degree vs excitation energy. The latter dependences were measured at fixed PL intensity, so we had to vary the laser power. Insofar as the PL intensity can be a measure of the excitonic generation rate, a dependence of this kind seems the most pure sensor of the role of the kinetic energy of the photocreated excitons.

It can be seen from Fig. 5 that, on the one hand, a noticeable heating occurs at a very small energy excess of exciting quanta above the bottom of the excitonic band. [In fact, the excess of 2 meV in Fig. 5(a) corresponds to excitation into the low-energy side of the excitonic line.] That is, a heating process can occur in which the kinetic energy of the excitons is relatively unimportant (e.g., heating due to non-radiative exciton recombination, for which the recombination energy is much greater than the kinetic energy). On the other hand, when the energy excess is increased, the PD range in which the heating occurs does narrow itself noticably [Fig. 5(a)], while the polarization decreases manifesting the temperature increase [Fig. 5(b)]. That is, another contribution to the heating effect also exists which may be due to cooling of hot excitons. We note that the magnitude of the energy-dependent part of the heating depends on the QW width: no effect of energy excess on polarization degree was observed in the same sample in the 100 Å QW, while in the 40 Å QW this effect was even more pronounced.

In support of the phononic origin of the heating effect, we assumed that the lattice temperature obeys the solution of the one-dimensional heat-conductance problem for the Debye limit, so that $G \propto (T^4 - T_0^4)$, where G is pump density (equal to the heat flux in steady state), and, since polarization varies as T^{-1}, we fitted the ''polarization vs pump'' dependences with a $T^{-1}(G)$ law. A good qualitative agreement is obtained in this way everywhere [see Figs. 1, 4, 5(a)].

In conclusion, we believe that the effect of the photo-induced heating of Mn spins under usual conditions of the magneto-luminescence experiments is rather common with diluted magnetic semiconductors and quantum structures, either doped or undoped. We have shown that the effect can often be explained by crystal lattice heating rather than by heating via a 2DEG. Can the spin-system of magnetic ions be taken out from thermal equilibrium with the lattice by photo-excited excitons? We think yes, but for the more dilute magnetic layers.[10] Such a dynamic depolarization effect (which is not heating in the true sense of the word) will then coexist with the overall heating studied here, because the latter is still not expected to vanish. Some of the experiments suggested in the present paper may be useful for segregation of the mechanisms.

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[1] B. König, I.A. Merkulov, D.R. Yakovlev, W. Ossau, S.M. Ryabchenko, M. Kutrowski, T. Wojtowicz, G. Karczewski, and J. Kossut, Phys. Rev. B **61**, 16 870 (2000).

[2] S.M. Ryabchenko, Yu.G. Semenov, and O.V. Terletskii, Zh. Éksp. Teor. Fiz. **82**, 951 (1982) [Sov. Phys. JETP **55**, 557 (1982)].

[3] S.I. Gubarev, T. Ruf, and M. Cardona, Phys. Rev. B **43**, 14 564 (1991).

[4] D. Wolverson, S.V. Railson, M.P. Halsall, J.J. Davies, D.E. Ashenford, and B. Lunn, Semicond. Sci. Technol. **10**, 1475 (1995).

[5] A.V. Koudinov, Yu.G. Kusrayev, B.P. Zakharchenya, and V.N. Yakimovich, Fiz. Tverd. Tela **37**, 660 (1995) [Phys. Solid State **37**, 359 (1995)].

[6] V.F. Aguekian, D.E. Ashenford, B. Lunn, A.V. Koudinov, Yu.G. Kusrayev, and B.P. Zakharchenya, Phys. Status Solidi B **195**, 647 (1996).

[7] Yu.G. Kusrayev and A.V. Koudinov, Phys. Status Solidi B **190**, 315 (1995).

[8] In the same experimental conditions we observed, as in Ref. 1, the effect of heating on the Zeeman splitting of luminescence line, however, for the sake of convenience we used the PL polarization measurements.

[9] A.V. Scherbakov, A.V. Akimov, D.R. Yakovlev, W. Ossaw, L.M. Mollenkamp, S. Tatarenko, and J. Cibert, Solid State Commun. **120**, 17 (2001).

[10] D. Keller, D.R. Yakovlev, B. König, W. Ossau, Th. Gruber, A. Waag, L.W. Mollenkamp, and A.V. Scherbakov, Phys. Rev. B **65**, 035313 (2002).