Inversion of magnetic hysteresis in semimagnetic superlattices

V. P. Kochereshko, G. R. Pozina, I. A. Merkulov, and D. R. Yakovlev
Physicotechnical Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia

G. Landwehr, W. Ossau, and A. Waag
University of Wurzburg 97074 Wurzburg, Germany

(Submitted 24 January 1995)
Pis'ma Zh. Éksp. Teor. Fiz. **61**, No. 5, 390–393 (10 March 1995)

A sign reversal of the magnetic hysteresis has been observed in CeTe/CdMnTe superlattices when the sample is excited by unpolarized light in the region of interband transitions. This effect stems from a nonequilibrium magnetization of manganese ions during spin relaxation of photogenerated carriers at Zeeman sublevels in the magnetic field. © *1995 American Institute of Physics.*

In this letter we are reporting a study of $CdTe/Cd_{1-x}Mn_xTe$ superlattices grown by molecular beam epitaxy with different barrier compositions: $x=0.1$ and 0.25. The period of the superlattice was varied from 24 to 120 Å ; the thickness of the barriers was equal to the width of the wells.

Figure 1a shows the Zeeman splitting of the exciton levels found from an analysis of reflection spectra in a magnetic field in the Faraday geometry[1] for $CdTe/Cd_{0.9}Mn_{0.1}Te$ superlattices with a period 40 Å . The saturation on this plot reflects the complete alignment of free Mn spins along the field. This is not to say that there is a complete alignment of all the magnetic ions of the sample, since at $T=1.6$ K a large fraction of the manganese ions are in antiferromagnetic clusters, whose magnetic moments are smaller by a large factor than the sum of the moments of the constituent particles. The antiferromagnetic order of the manganese ions in neighboring sites of the crystal lattice begins to be disrupted in fields of about 10 T, at which the paramagnetic component of the magnetization reaches saturation.

To measure the orientation of the magnetic moments of the carriers, we studied the circular polarization of the exciton photoluminescence which arises in a magnetic field in the Faraday geometry (Fig. 1b). In weak fields ($H<0.1$ T), the degree of circular polarization $\rho_c(H)$ is proportional to the external magnetic field H and to the average magnetization of the manganese ions, $\langle M\rangle$. It is the same, within a constant factor, as the magnetization curve of the spin system of manganese ions. The reason for the early onset of saturation on the $\rho_c(H)$ curves in fields $H>0.2$ T is that in such fields the energy of the Zeeman splitting is much larger than kT (at $T=1.6$ K), and the spins of essentially all the electrons are oriented along the exchange field.

A study of the polarized photoluminescence revealed a hysteresis loop in the case of magnetization reversal. It turns out that the sign of this loop depends on the conditions under which the sample is illuminated. Figure 2a shows a magnetization-reversal curve

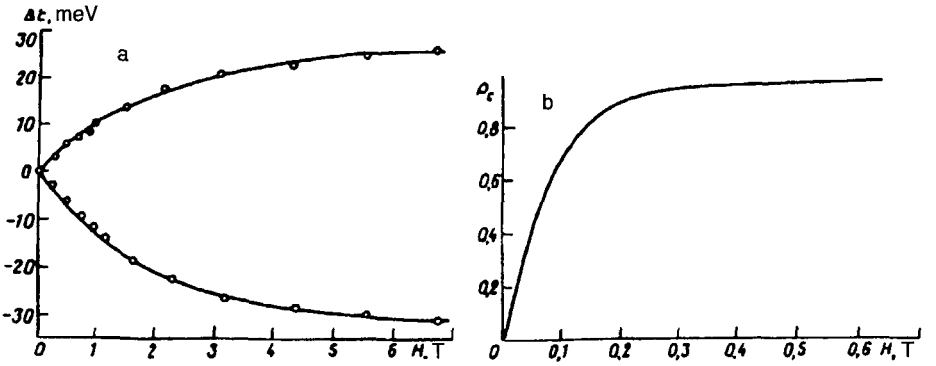

FIG. 1. a—Zeeman splitting of an exciton in $CdTe/Cd_{0.9}Mn_{0.1}Te$ superlattices in a magnetic field oriented parallel to the axis of the superlattice; b—degree of circular polarization of the photoluminescence induced by a magnetic field in the Faraday geometry, $k \| H \| z$, where k is the wave vector of the light, and z is the superlattice axis.

for a sample measured during constant illumination by a 1 W/cm^2 He–Ne laser. Figure 2b shows a magnetization-reversal curve found in a more complex procedure: The magnetization reversal was carried out in darkness; the light was turned on only during the measurement of the circular polarization of the photoluminescence. Each of the curves (Fig. 2, a and b) exhibits a hysteresis: The magnetization branch $\rho_c^{\uparrow}$ does not coincide with the demagnetization branch $\rho_c^{\downarrow}$. In a dark experiment, the magnetization curve $\rho_c^{\uparrow}$ runs below the demagnetization curve $\rho_c^{\downarrow}$, as is typical of magnetic hysteresis.[2] In the case of magnetization reversal in light, these curves trade places (Fig. 2a). The nonequilibrium remanent magnetization disappears over a time scale of 10^2–10^3 s. Consequently, during the magnetization-reversal cycle, energy is lost in darkness, while in light there is an evolution of energy equal to the area of the corresponding hysteresis loop,

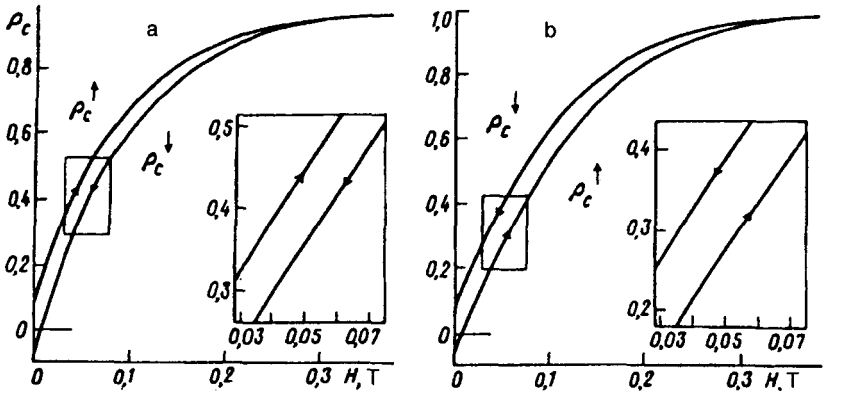

FIG. 2. Hysteresis in the degree of polarization of the photoluminescence during magnetization reversal of the sample. a—In light; b—in darkness.

$E = \oint M dH$. The energy evolved is evidently transferred to the system of photogenerated carriers.

In further experiments we focused on determining the conditions for the onset of a negative remanent magnetization. It turned out that if the field was increased to 5–6 T after or during the illumination, and if a demagnetization was then carried out in darkness, then the memory of the illumination would be "erased." In other words, the remanent magnetization would be positive (parallel to the external field). If the field was reduced under conditions of constant illumination, on the other hand, the sign of the remanent magnetization turned out to be negative. To eliminate an uncertainty in the results of the measurements of the dark hysteresis due to the history of the sample, we placed the sample in a strong field $H = 5$ T before each measurement cycle. The strong field was later turned off in darkness. A sample prepared in this manner was immersed in a weak magnetic field, illuminated for a certain time t, and demagnetized in darkness to $H = 0$. We then determine the sign and magnitude of the remanent magnetization from the degree of circular polarization of the photoluminescence. It turned out that the typical illumination times required for the onset of a negative remanent magnetization depend on the strength of the magnetic field during the illumination. For illumination in a field $H < 0.1$ T, there is essentially no negative magnetization. In fields of 0.3–0.5 T, an illumination time on the order of 100 s is required for the onset of a negative magnetization. Finally, at $H > 0.5$ T, the effect arises essentially instantaneously. The magnitude of the negative remanent magnetization falls off to zero as the field is increased to 2–3 T during illumination.

The long times (10^2–10^3 s) required for the disappearance of the magnetization of the electron spins in a field $H = 0$ may be due to the onset of a strong correlation among the spins of a large number of magnetic ions.[2-4] At $T = 1.6$ K, such clusters arise because of an exchange interaction between manganese ions which occupy positions in the first and second coordination spheres of the Cd sublattice with respect to each other. The exchange-interaction constants of the nearest neighbors and the next-nearest neighbors in the Heisenberg Hamiltonian $H = -2J(S_1 S_2)$ are $J_1 k_B = -6.1$ K and $J_2/k_B = -1.1$ K, respectively. Let us assume that the slowly relaxing part of the magnetization stems from the moments of clusters whose magnetization reversal involves the surmounting of high activation barriers. The relaxation time of their magnetization is therefore given by $\tau = \tau_0 \exp(\epsilon_\alpha / k_B T)$. In our case, the value of this relaxation time would be 10^2–10^3 s. Here ϵ_α is the activation energy, and τ_0 is a time scale which may be associated with magnetic-dipole interactions. In this case we would have, in order of magnitude, $\tau_0 \propto \hbar a^3 / M^2 \propto 10^{-10}$ s. Here M is the total magnetic moment of a manganese ion, which is $5\mu_B$, and $a \propto 6.5 \times 10^{-8}$ cm is the lattice constant. The activation energy is then $\epsilon_\alpha \propto k_B T \ln(\tau/\tau_0) \propto 3.5$–$4$ meV. In the absence of a magnetic field, a change in the direction of all spins of a cluster and thus a change in the sign of the magnetic moment of the cluster do not alter the energy of the system. The nonequilibrium magnetization thus vanishes over a time τ. The magnetic field splits the degenerate levels, and the equilibrium magnetization becomes nonzero. If ϵ_α does not change significantly in the process, the time scale for relaxation to equilibrium is again 10^2–10^3 s. It is natural to suggest that the activation energy for a state with magnetic moment directed opposite the field decreases by an amount $M_\Sigma H$, where M_Σ is the resultant moment of the cluster. In a field

of several teslas, the time scale for relaxation to equilibrium thus decreases by a factor of $\exp[(M_\Sigma H)/k_B T]$, and even for clusters with a single unpaired spin this time is about a second. Under the experimental conditions, the onset of the polarization would be perceived as instantaneous.

What happens when the sample is illuminated? Let us assume that the carriers undergo energy relaxation quickly and slide down to the bottom of the conduction band and the valence band unpolarized. In a weak magnetic field, the Zeeman splittings of exciton states are small, and the energy evolved in the establishment of equilibrium distribution of particles with respect to magnetic sublevels is not sufficient to activate the magnetization reversal of high-coercivity clusters. As the Zeeman splitting increases, the energy transferred to the spin system of the manganese in the course of flip–flop transitions increases; at a field of only 0.5 T in order of magnitude (Fig. 1a) it becomes comparable to the height of the activation barrier. This energy is comparable in order of magnitude to the energy of the exchange interaction between neighboring manganese ions. Under these conditions the spin transferred from excitons to the spin system of the magnetic ions may lead to a magnetization reversal of not only the paramagnetic fraction of the manganese ions but also the high-coercivity clusters. As a result of this "Overhauser effect,"[5] the slowly relaxing magnetic moments turn out to be oriented opposite the magnetic field. After dynamic equilibrium is reached, a long-lived remanent magnetization , which is directed opposite the magnetic field, arises. This remanent magnetization is manifested in the onset of a negative hysteresis loop. As we move to stronger fields, and the relaxation time of the high-coercivity clusters decreases rapidly, the negative remanent magnetization should disappear after dark demagnetization; this is what is observed experimentally. The reason for the onset of a negative hysteresis loop (a negative remanent magnetization) is thus an Overhauser effect involving antiferromagnetic clusters with a long spin-relaxation time. Although the model used here is extremely crude, the mechanism proposed here for the change in the sign of the hysteresis loop looks fairly unambiguous to us.

In conclusion we wish to thank the Volkswagen Foundation for support of this study.

[1]E. L. Ivchenko, A. V. Kavokin, V. P. Kochereshko *et al.*, Phys. Rev. B **46**, 7713 (1992).
[2]K. Binder A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).
[3]J. V. Hong, D. D. Awshalom *et al.*, J. Appl. Phys. **63**, 3285 (1988).
[4]D. D. Awshalom, J. V. Hong, L. L. Chang *et al.*, Phys. Rev. Lett. **59**, 1733 (1987).
[5]A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon Press, Oxford, 1978).

Translated by D. Parsons