Proc. XXXVII International School of Semiconducting Compounds, Jaszowiec 2008

MnAs Nanocrystals Embedded in GaAs

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Magnetic properties of MnAs nanocrystals embedded in GaAs are analyzed in the frame of phenomenological model proposed by Sasaki for ferritin superparamagnets. Our calculations explain qualitatively experimental data of magnetization versus temperature, obtained according to zero-field-cooled and field-cooled protocols. They show dynamics of magnetization of MnAs nanocrystals in range of temperature from 10 K to 320 K. There is transition from state in which very slow dynamics is observed (frozen state) to state in which dynamics is fast (quasi-superparamagnetic state).

PACS numbers: 75.20.–g, 75.75.+a

1. Introduction

Magnetic properties of small single domain particles is a topic developed since about fifties of 20th century (see for example [1]). Self-organized Ga(Mn)As nanocrystals embedded in gallium arsenide are example of that type of materials. Composites containing Mn ions are recognized as very interesting and promising spintronic materials, integrating magnetic and semiconductor features. Their advantages include Mn ferromagnetism above room temperature and GaAs compatibility with existing III–V technology. In this work macroscopic magnetic properties of such system, consisting of magnetic MnAs nanocrystals embedded in GaAs lattice, are explained in the frame of a model proposed by Sasaki for ferritin superparamagnets [2].

2. Samples and experiment

It is well known that Ga(Mn)As nanocrystals are formed during post-growth annealing of $Ga_{1-x}Mn_xAs$ layers grown by MBE technique on semi-insulating

GaAs (e.g. [3–5]). In our study we used a series of samples having Mn content, $x = 3$–5% and annealed at temperatures in the range of 500–600°C. More details of the growth procedure are given in Refs. [5, 6].

Transmission electron microscopy (TEM) experiments showed in our samples formation of two types of inclusions: (1) hexagonal MnAs nanocrystals of NiAs structure, and (2) cubic Ga(Mn)As nanocrystals of zinc-blende structure, which were usually much smaller than MnAs ones. TEM images revealed a wide distribution of MnAs nanocrystal sizes in each sample. The mean value of precipitate diameter varied from sample to sample in the range of 4 up to 16 nm.

Magnetization measurements, performed according to zero-field-cooled (ZFC) and field-cooled (FC) protocols, revealed at low temperatures a substantial difference between ZFC and FC data. It is illustrated for a chosen sample in Fig. 1. One can see for ZFC data a characteristic maximum at about

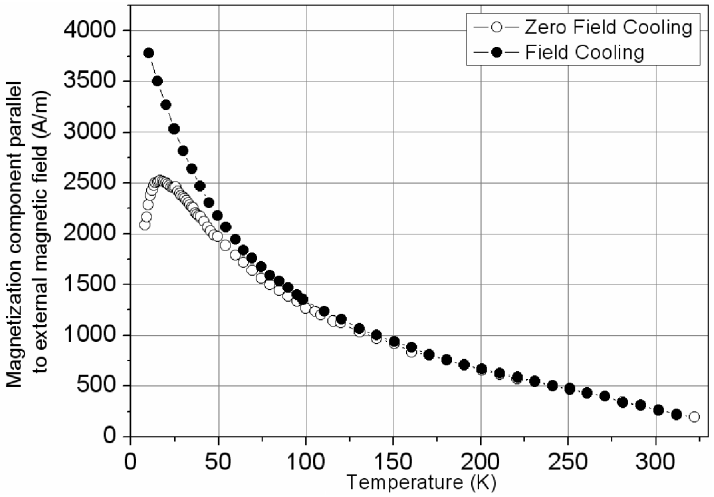

Fig. 1. Typical magnetization of a chosen sample for ZFC/FC protocols. External magnetic field was 0.004 T.

$T_{\max} \approx 17$ K, while FC data decreases monotonically with increasing temperature and for $T > T_{\max}$ it approaches gradually the ZFC points. For higher temperatures, above so-called blocking temperature T_{b}, ZFC magnetization curve overlaps the FC curve. T_{b} depicts a phase transition between ferromagnetic "frozen" state and quasi-superparamagnetic state. We found that the value of blocking temperature increased with increase in the mean size of MnAs nanocrystals, and for the measured set of samples it varied from about 100 K up to about room temperature.

3. Phenomenological model of the system

To explain the experimental data we adopted a phenomenological model introduced for ferritin superparamagnets by Sasaki et al. [2]. We assumed

as follows:

- Each MnAs nanocrystal is a single magnetic domain having dipolar magnetic moment, μ only. At a given temperature T, and in an external magnetic field (B_0), magnetic moment of an individual nanocrystal can occupy one of two states with energies $\pm\mu B_0$.
- These states are separated by an energy barrier (E_b), which is related to presence of magnetocrystalline anisotropy and magnetic dipolar interactions.
- The barrier is proportional to the volume of given nanocrystal. Proportionality coefficient, called effective barrier energy density is denoted as $K_{\mathrm{eff\,DC}}$.
- Characteristic frequency of magnetic moment thermal reversing f_0 is independent of temperature and properties of nanocrystal.
- Master equation for initial conditions, $T(t)$ and $B_0(t)$ taken from an experiment, is as follows: $\frac{\mathrm{d}}{\mathrm{d}t}p_1(t) = -W_{12}(t)p_1(t) + W_{21}(t)[1 - p_1(t)]$, where $p_1(t)$ denotes an occupation probability of state 1, while $p_2(t) = 1 - p_1(t)$ denotes an occupation probability of state 2.

  Transition rates have the following form:

$$W_{12}(t) = \frac{1}{2} f_0 \exp\left(\frac{-K_{\mathrm{eff\,DC}}V - \mu B_0(t)}{k_\mathrm{B}T(t)}\right),$$

$$W_{21}(t) = \frac{1}{2} f_0 \exp\left(\frac{-K_{\mathrm{eff\,DC}}V + \mu B_0(t)}{k_\mathrm{B}T(t)}\right).$$

Magnetization of the system is given by

$$M(t) = [2p_1(t) - 1]\mu n,$$

where n denotes concentration of nanocrystals in unit of volume. From TEM experiments we estimated $n = 7.2 \times 10^{22}$ m^{-3}. *We assumed that our system is monodispersive, i.e. all nanocrystals are of the same volume*, $V = 5 \times 10^{-25}$ m^3 (an average diameter of the nanocrystal equal to about 10 nm) with $\mu = 1000\mu_\mathrm{B}$. In our calculations we used $K_{\mathrm{eff\,DC}} = 5 \times 10^4$ J/m^3, which is typical value reported in literature of superparamagnetic systems (e.g. [7–9]). Results of calculations within proposed model are shown in Fig. 2.

4. Discussion

Our calculations reproduce qualitatively experimental data for a sample shown in Fig. 1. In this case at low temperatures, i.e. below 81 K, when $E_\mathrm{b} \gg k_\mathrm{B}T$, the thermal relaxation time is very long with respect to duration of experiment. System needs a lot of time to approach thermal equilibrium, therefore magnetization measurements depict some "frozen" state of the sample, which depends on cooling conditions. For higher temperatures, when $E_\mathrm{b} \ll k_\mathrm{B}T$, the system reaches its thermal equilibrium very quickly because its relaxation time becomes much

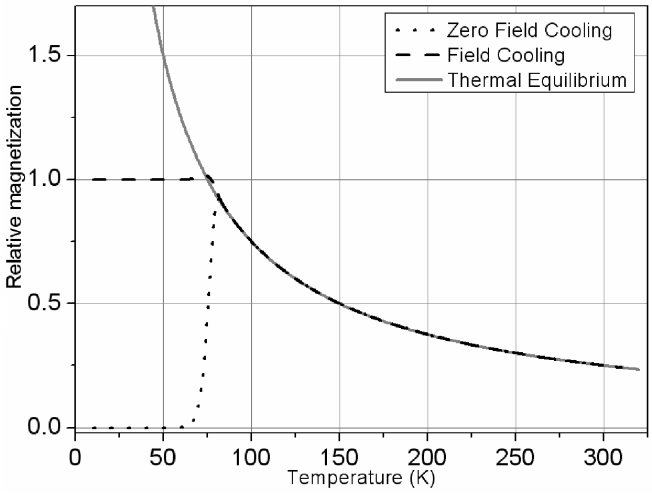

Fig. 2. Results of calculations for monodispersive system.

shorter than the observation time. In this case magnetization does not depend on cooling "history" of the sample. This behavior is very similar to superparamagnetism and we call it "unblocked" state.

In this model of monodispersive system transition between ferromagnetic "frozen" state and "unblocked" state at 81 K is very sharp, because in this case energy barrier has the same height for all nanocrystals. To explain quantitatively the experimental data, more advanced model should be developed which takes into account a wide distribution of the nanocrystals volume.

5. Conclusions

Our results show dynamics of magnetization of MnAs nanocrystals in the range of temperatures from 10 K to 320 K. There is a transition from state in which very slow dynamic is observed (frozen state) to state in which dynamic is fast (quasi-superparamagnetic state). There are two known reasons of such behavior:

- magnetocrystalline anisotropy of nanocrystals,
- magnetic interactions between nanocrystals.

Acknowledgments

We thank Faculty of Materials Science and Engineering of Warsaw University of Technology for providing transmission electron microscopy. We appreciate Prof. Anna Ślawska-Waniewska and Dr. hab. Piotr Kossacki for helpful discussions. This work was partially supported by European Commission under grant No. MTKD-CT-2005-029671 and by the Polish Ministry of Science and High Education by grant No. N 202 052 32/1189.

References

[1] C.P. Bean, J.D. Livingston, *J. Appl. Phys.* **30**, 120S (1959).

[2] M. Sasaki, P.E. Jönsson, H. Takayama, H. Mamiya, *Phys. Rev. B* **71**, 104405 (2005).

[3] J. De Boeck, R. Oesterholt, A. Van Esch, H. Bender, C. Bruynseraede, C. Van Hoof, G. Borghs, *Appl. Phys. Lett.* **68**, 2744 (1996).

[4] M. Moreno, A. Trampert, B. Jenichen, L. Däweritz, K.H. Plogg, *J. Appl. Phys.* **92**, 4672 (2002).

[5] A. Kwiatkowski, D. Wasik, M. Kamińska, R. Bożek, J. Szczytko, A. Twardowski, J. Borysiuk, J. Sadowski, J. Gosk, *J. Appl. Phys.* **101**, 113912 (2007).

[6] M. Adell, L. Ilver, J. Kanski, V. Stanciu, P. Svedlindh, J. Sadowski, J.Z. Domagala, F. Terki, C. Hernandez, S. Charar, *Appl. Phys. Lett.* **86**, 112501 (2005).

[7] S. Tomita, P.E. Jönsson, K. Akamatsu, H. Nawafune, H. Takayama, *Phys. Rev. B* **76**, 174432 (2007).

[8] J.-O. Andersson, C. Djurberg, T. Jonsson, P. Svedlindh, P. Nordblad, *Phys. Rev. B* **56**, 13983 (1997).

[9] E. Vincent, Y. Yuan, J. Hammann, H. Hurdequint, F. Guevara, *J. Magn. Magn. Mater.* **161**, 209 (1996).