

# Polarized neutron reflectivity of dilute magnetic semiconductors

Amitesh Paul\*, Heiko Braak, Daniel E. Bürgler, Reinert Schreiber, Diana Rata, Peter Grünberg, Claus M. Schneider, Thomas Brückel

*Institut für Festkörperforschung, Forschungszentrum Jülich GmbH, D-52425 Jülich, Germany*

## Abstract

We report on a polarized neutron reflectivity investigation of the magnetization in Ge-based dilute magnetic semiconductors. We could observe a net magnetization from the splitting of the non-spin flip reflectivity patterns, which measure the magnetic moment parallel and antiparallel to the applied field. This contrast is visible at 50 K, at remanence and it is pronounced at higher fields even at 250 K for an inhomogeneous specimen. For a homogeneous sample the magnetic variation is visible only at 50 K and above 1.0 kOe. Thus, polarized neutron reflectivity can be a useful tool for investigating the magnetism in homogeneous and inhomogeneous thin film magnetic semiconductors.

© 2007 Elsevier B.V. All rights reserved.

PACS: 61.12.Ha; 75.50.Pp

Keywords: Polarized neutron reflectometry; Magnetic semiconductors

In recent years one of the remarkable achievements in spintronics was the observation of hole-mediated ferromagnetism in III–V compounds [1]. However, the growth of group-IV ferromagnetic semiconductors such as  $\text{Mn}_x\text{Ge}_{1-x}$  has also triggered strong interest in searching for higher ordering temperatures  $T_c$  in Ge-based compounds [2]. Recently, we have reported [3] on the successful synthesis of a new candidate for a dilute magnetic semiconductor (DMS) compound: an alloy of Ge, Fe, and Mn. The addition of Fe to MnGe enables us to conserve epitaxial growth for higher total concentrations of magnetic species, which results in an increase of  $T_c$ . The alloy films are prepared by growing repeatedly Ge/Mn/Fe trilayers with various thicknesses of the Ge base layers. When the Ge thickness becomes comparable with the diffusion length of Mn and Fe in Ge, a homogeneous material without interfaces is formed as confirmed by X-ray reflectivity (XRR). Here, we consider two samples with different compositions:

$[\text{Ge}(40 \text{ \AA})/\text{Fe}(4 \text{ \AA})/\text{Mn}(2 \text{ \AA})]_{\times 11}/\text{ZnS}(500 \text{ \AA})$  named “DMS-79” and

$[\text{Ge}(40 \text{ \AA})/\text{Fe}(2 \text{ \AA})/\text{Mn}(4 \text{ \AA})]_{\times 11}/\text{ZnS}(500 \text{ \AA})$  named “DMS-80”.

The samples were grown by molecular beam epitaxy. Details have been given elsewhere [3,4]. Fig. 1 shows the XRR patterns for a typical multilayer sample with different annealing temperatures  $T_A$ . For  $T_A = 300^\circ\text{C}$  a homogeneous alloy is observed. Magnetization measurements by a superconducting quantum interference device (SQUID) and magnetotransport measurements reveal ferromagnetic

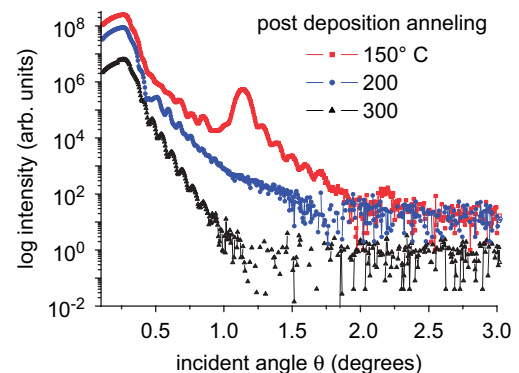


Fig. 1. (Color online.) XRR data for “DMS-79” at different annealing temperatures. For  $T_A > 200^\circ\text{C}$  the multilayer Bragg peaks disappear.

\*Corresponding author. Tel.: +49 2461 613141; fax: +49 2461 612610.  
E-mail address: [A.Paul@fz-juelich.de](mailto:A.Paul@fz-juelich.de) (A. Paul).

ordering, but show for “DMS-80” two different  $T_c$ s at 205 K and above 300 K, which we relate to two magnetic phases [4]. Transmission electron microscopy (TEM) indeed shows the formation of precipitates depending on the preparation condition and the content of magnetic species. Interestingly, “DMS-79” does not show the high- $T_c$  phase and appears in the TEM picture to be fairly homogeneous compared to the other samples. An unequi-

vocal assignment of the magnetic phases to the precipitates and the matrix, respectively, requires more knowledge about the magnetic structure.

The hysteresis loops of “DMS-79” and “DMS-80” measured at different temperatures are shown in Fig. 2. Polarized neutron reflectivity has been used earlier to study DMS compounds based on GaAs [5,6]. Ferromagnetic behavior in DMS is often inconclusive due to the

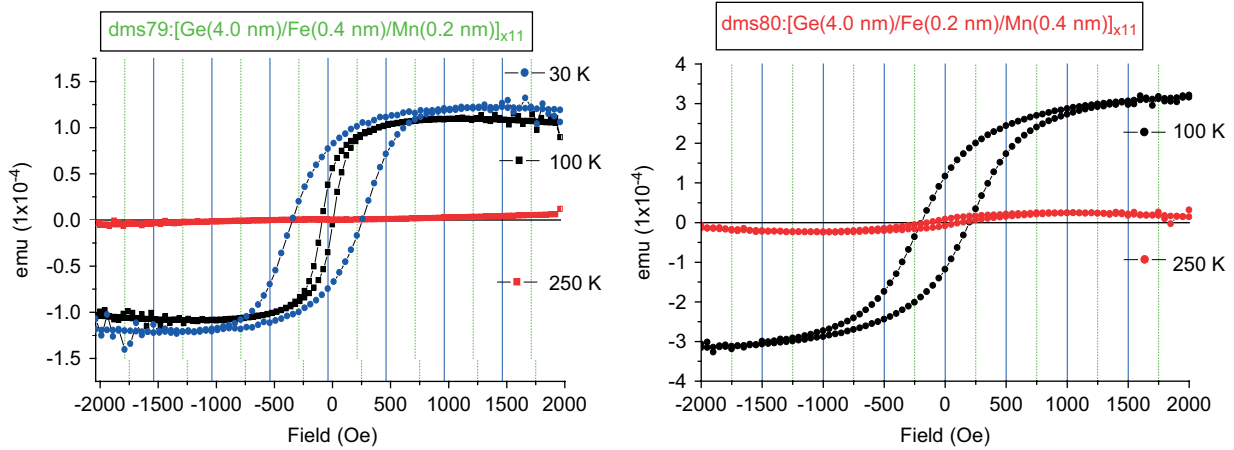


Fig. 2. (Color online.) Hysteresis loops from “DMS-79” and “DMS-80” taken at different temperatures.

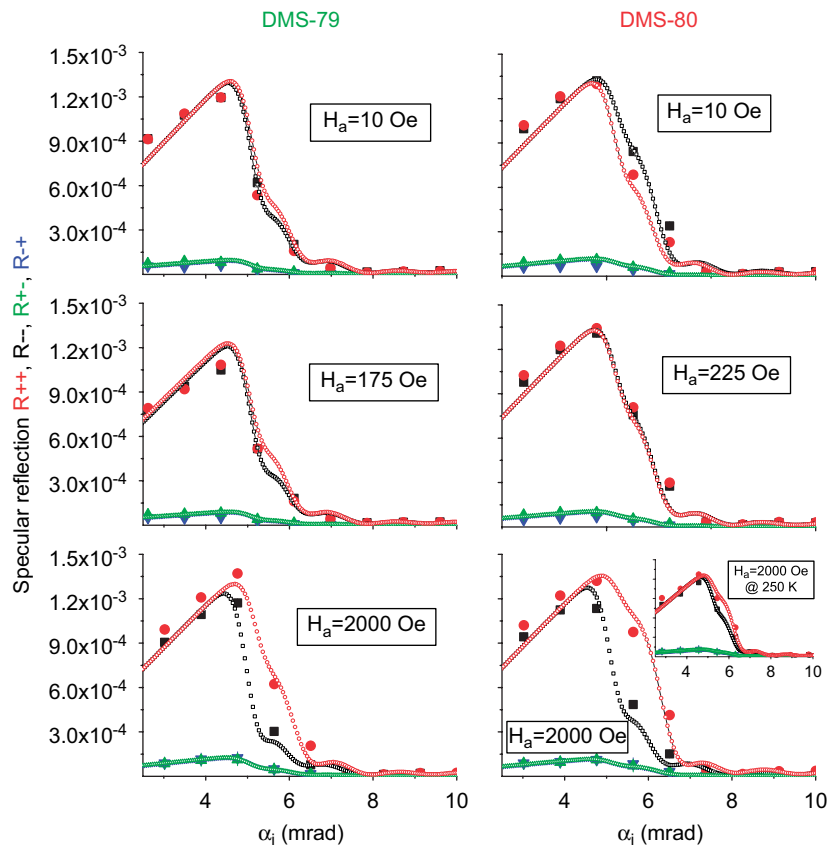


Fig. 3. (Color online.) Measured (solid symbols) and fitted (open circle) NSF [ $R_{--}$  (black square) and  $R_{++}$  (red circle)] and SF [ $R_{+-}$  (green triangle) and  $R_{-+}$  (blue downtriangle)] reflectivity patterns of “DMS-79” and “DMS-80” measured at different applied fields  $H_a$  and at 50 K. The inset of the last panel shows data for “DMS-80” taken at 250 K. No SF signal can be observed as can be derived from the comparison with the saturated state at 2.0 kOe, where all SF intensities are due to non-ideal polarization only.

formation of isolated domains. Here we investigate homogeneous (DMS-79) and inhomogeneous (DMS-80) specimens based on Ge which is interesting technologically and intrinsically.

Polarized neutron reflectometry is performed on the HADAS reflectometer at FRJ-2 at a wavelength of 4.52 Å. Fig. 3 shows measurements for both specimens, at 50 and 250 K, and at various applied fields. The specular reflectivities in the spin-flip (SF) channels ( $R_{+-}$  and  $R_{-+}$ ) correspond to in-plane magnetization components perpendicular to the guiding field  $H_a$ , whereas the non-spin-flip (NSF) channels ( $R_{++}$  and  $R_{--}$ ) yield information about the magnetization component collinear to  $H_a$ . Here + and – signs to the intensity  $R$  represent the polarization parallel and antiparallel to the guiding field.

A splitting of the NSF reflectivity channels can be observed for both the specimens at an applied field larger than the saturation field (e.g. at  $H_a = 2000$  Oe). This splitting indicates a significant contrast between magnetic moments parallel and antiparallel to  $H_a$ . It is visible only at 50 K and above 1.0 kOe for “DMS-79”. Similar contrast is seen for “DMS-80” even at somewhat lower fields at 50 K, and this net magnetization is pronounced at higher fields even at 250 K (inset). At 50 K, measured at 10 Oe, “DMS-80” shows very small splitting of the  $R_{++}$  and  $R_{--}$  which is opposite to that shown for fields above the reversal field (225 Oe). This along with no true SF signal indicates a spontaneous ferromagnetism which can be attributed to

the clustering in the sample. No such splitting at remanence is noticed for a homogeneous “DMS-79”. Unfortunately, no resolvable scattering intensities from the individual elements of the samples could be seen, and the data have been fitted considering a mixed layer of Ge, Fe, and Mn capped with ZnS. Even though these are very difficult materials to measure with polarized neutrons due to the relatively low-magnetic ion concentration, the magnetic signal seen here is promising. A brighter neutron source yielding higher flux and less background is expected to provide more insight into the magnetism of diluted ferromagnetic semiconductors. Corresponding experiments are planned for the future.

## References

- [1] H. Ohno, *J. Magn. Magn. Mater.* 200 (1999) 110.
- [2] Y.D. Park, A.T. Hanbicki, S.C. Erwin, C.S. Hellberg, J.M. Sullivan, J.E. Mattson, T.F. Ambrose, A. Wilson, G. Spanos, *Science* 295 (2002) 651.
- [3] H. Braak, R. Gareev, D.E. Bürgler, R. Schreiber, P. Grünberg, C.M. Schneider, *J. Magn. Magn. Mater.* 286 (2005) 46.
- [4] H. Braak, A.D. Rata, D.E. Bürgler, P.A. Grünberg, C.M. Schneider, to appear.
- [5] H. Kępa, J. Kutner-Pielaszek, A. Twardowski, C.F. Majkrzak, J. Sadowski, T. Story, T.M. Giebultowicz, *Phys. Rev. B* 64 (2001) 121302.
- [6] B.J. Kirby, J.A. Borchers, J.J. Rhyne, S.G.E. te Velthuis, A. Hoffmann, K.V. ODonovan, T. Wojtowicz, X. Liu, W.L. Lim, J.K. Furdyna, *Phys. Rev. B* 69 (2004) 081307.