Available online at www.sciencedirect.com
ScienceDirect
Physica B 397 (2007) 36–42
PHYSICA B
www.elsevier.com/locate/physb

Ferromagnetic semiconductor superlattices studied by polarized neutron reflectometry

H. Kępa[a,b,*], C.F. Majkrzak[c], A. Yu. Sipatov[d], T.M. Giebultowicz[b]

[a]Institute of Experimental Physics, Warsaw University, ul. Hoża 69, 00-681 Warszawa, Poland
[b]Physics Department, Oregon State University, 301 Weniger Hall, Corvallis, OR 97331, USA
[c]NIST Center For Neutron Research, National Institute of Standards and Technology, Gaithersburg, MD 20899, USA
[d]Kharkov State Polytechnic University, 21 Frunze St., Kharkov 310002, Ukraine

Abstract

The results of extensive neutron reflectometry studies of EuS-based ferromagnetic semiconductor superlattices (SL's) which exhibit interlayer exchange coupling (IEC) are presented. The SL's grown on two different substrates, (0 0 1)KCl and (1 1 1)BaF_2, with two types of nonmagnetic spacer material, PbS (narrow gap) and YbSe (wide gap semiconductor), were investigated. Neutron reflectometry measurements revealed pronounced antiferromagnetic IEC in the systems grown on (0 0 1)KCl substrate, whereas in the EuS/PbS superlattices grown on (1 1 1)BaF_2 a ferromagnetic IEC has been found. The in-plane magnetic anisotropy and the population of the magnetic domains were studied by polarized neutron reflectivity. The in-plane directions of easy-axes has thus been determined in the all investigated systems. A nonuniform distribution of domains magnetization directions among the easy in-plane axes has been found.
© 2007 Elsevier B.V. All rights reserved.

PACS: 75.25.+z; 75.50.Pp; 68.65.Cd; 61.12.Ha; 61.12.Ld

Keywords: Magnetic semiconductor; Superlattices; Polarized neutron reflectivity

1. Introduction

With the advent of "spintronics", ferromagnetic semiconductor superlattices, and the magnetic interlayer exchange interactions (IEC) within them, have played a crucial role both from a practical and fundamental research point of view. In the all-semiconductor EuS/PbS and EuS/YbSe superlattices grown on (0 0 1)KCl substrates, a pronounced antiferromagnetic (AFM) IEC has been observed by SQUID magnetometry [1] and neutron scattering [2]. The AFM alignments of the magnetization vectors in the adjacent EuS layers were clearly seen in neutron reflectometry experiments even for a very thick (up to 400 Å) PbS spacer. Unfortunately, the very low Curie temperature of EuS ($T_C = 18$ K) makes this material unsuitable for practical applications. So, the search for above room temperature semiconductor ferromagnets is of primary importance for spintronics. Recent progress in the MBE growth of III–V-type diluted magnetic semiconductors (DMS) (such as GaMnAs, InMnAs, GaMnN) [3] raised new hope for a 'good' spintronics material. In particular, the strongly p-type InMnAs and GaMnAs exhibit the desired FM behavior. The Curie points of these materials are still much below room temperature, which limits their practicality. Nevertheless, both classes of material, EuS-based and GaAs-based DMS's, may play an important role in developing prototypes of future spintronic devices. Apart from IEC phenomena in SL's, of primary importance is the in-plane anisotropy and magnetic domain structure. The most desirable situation from the spintronics viewpoint is spontaneous formation of a single-domain FM state, thus reducing the need for an external magnetic field. Such a situation was indeed observed in our earlier polarized neutron reflectometry (PNR) studies of the GaMnAs/GaAs system [4]. Yet, our

*Corresponding author. Institute of Experimental Physics, Warsaw University, ul. Hoża 69, 00-681 Warszawa, Poland. Tel.: +48 22 5532197; fax: +48 22 6294229.
E-mail address: Henryk.Kepa@fuw.edu.pl (H. Kępa).

0921-4526/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.physb.2007.02.019

current insight into the domain structure of the new epitaxial ferromagnets is still lacking. Specular [5] and off-specular [6] PNR has recently been used to obtain closer insight into the lateral magnetic structure in thin films and SL's. Here, we review specular PNR studies that provide information about the in-plane magnetic anisotropy in the EuS layers in antiferromagnetically coupled EuS/PbS and EuS/YbSe SL's grown on (0 0 1)KCl substrates as well as in ferromagnetically coupled EuS/PbS SL's grown on (1 1 1) BaF_2 substrates.

2. Domain distribution in antiferromagnetically coupled EuS/PbS and EuS/YbSe SL's grown on (0 0 1)KCl

Typical unpolarized neutron reflectivity patterns for AFM coupled EuS-based superlattices are presented in Fig. 1. Although unpolarized neutron beams proved very useful in the studies of the IEC phenomena, they give no information about the in-plane magnetic moment orientations; for this the use of polarized neutrons is necessary.

The principles of neutron polarization analysis, laid out in Ref. [7], are summarized in Fig. 2. In most neutron polarization analysis experiments, four cross sections are measured, i.e., (++), (−−), (+−), and (−+). In the first two the spin state of the neutron does not change (non-spin-flip or NSF processes) whereas in the last two the neutron spin is reversed (spin-flip or SF processes).

In order to investigate the distribution of in-plane magnetization directions of magnetic domains, and their relative population, the sample has to be mounted with a specific crystallographic axis pointing in a given (usually, horizontal or vertical) direction. Then, a series of measurements have to be carried out with the sample rotated about the normal to its reflecting surface. The rotation changes the parallel and perpendicular components of the sample magnetization with respect to the neutron polarization direction that in turn is reflected in spin dependent neutron scattering intensities.

Here, we will discuss the results of such measurements for two different superlattices, EuS/PbS (35 Å/4 Å) and EuS/YbSe (44 Å/20 Å). In each case an orientation of the sample was found such that all spin-dependent SL Bragg peak intensities were equal; this is shown in Fig. 3. Such an equality is indicative of either a uniaxial domain sample or one with two 90° domains having all domains aligned along directions making angles of 45° with the neutron polarization direction. It was determined that in the case of EuS/PbS SL's the domains are oriented along in-plane ⟨2 1 0⟩-type directions, whereas in EuS/YbSe SL's magnetic domains were aligned along ⟨1 1 0⟩-type directions.

To see whether both [2 1 0] and [1 $\bar{2}$ 0] directions in EuS/PbS SL's are populated by magnetic domains, and what their relative population is, the sample was remounted with the [2 1 0] axis horizontal. A series of measurements were then carried out with the sample rotated about the normal to its reflecting surface by ±20° from this position. The

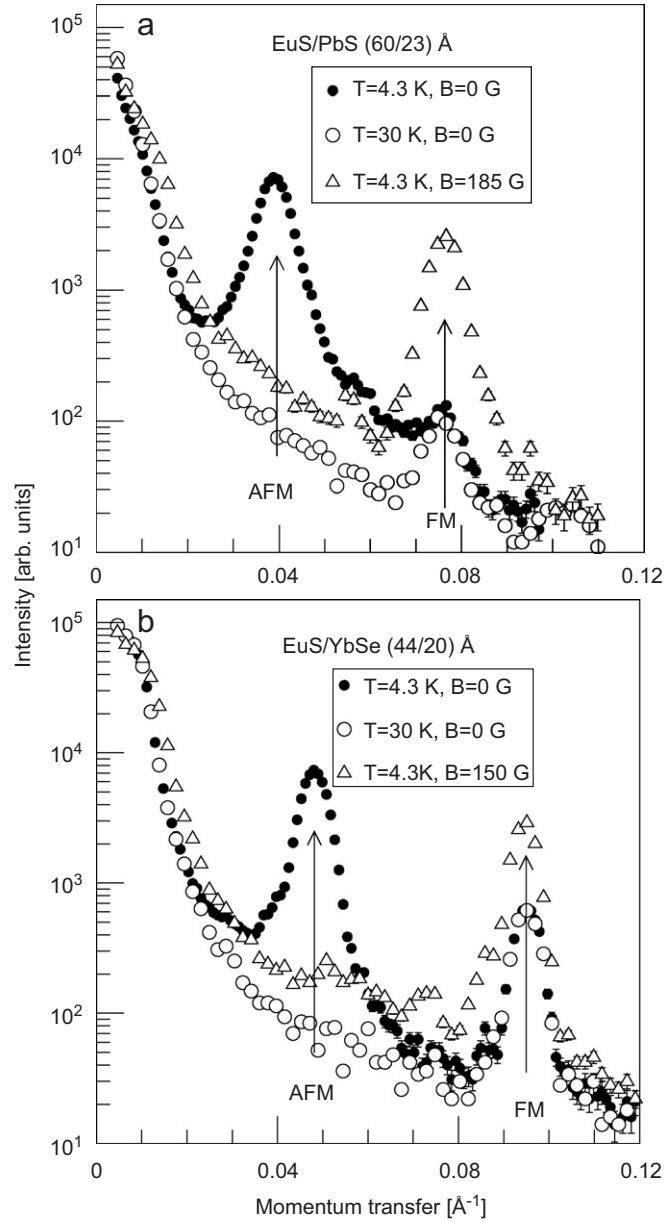

Fig. 1. Neutron reflectivity spectra for (a) EuS/PbS (60 Å/23 Å) × 10 and (b) EuS/YbSe (44 Å/20 Å) superlattices grown on (0 0 1)KCl substrates, measured below and above T_C in zero (filled and open circles, respectively) and in a saturating (triangles) magnetic field.

results are presented in Fig. 4(a). Analogous data obtained for a EuS/YbSe superlattice mounted with the [1 0 0] in-plane axis vertical are shown in Fig. 4(b).

To explain the observed effects it was assumed that the samples consist of two types of domains with their magnetizations aligned along two perpendicular directions, [1 1 0] and [1 $\bar{1}$ 0] in the case of the EuS/YbSe specimen, and [2 1 0] and [1 $\bar{2}$ 0] in the EuS/PbS sample. The area of the sample occupied by these domains is $S_1 = xS$ and $S_2 = (1-x)S$, respectively, where S is the total reflecting area of the sample (see Fig. 5). In the case of antiferromagnetically coupled SL's, the first SL Bragg peak is purely

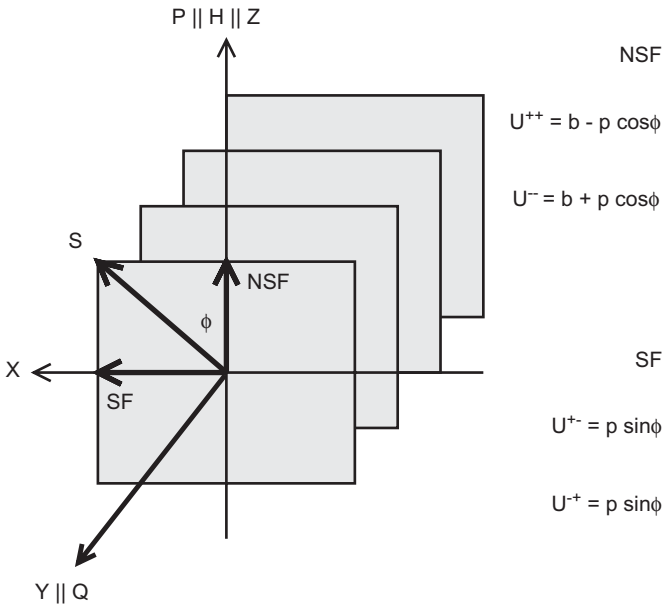

Fig. 2. The principles of neutron polarization analysis. Neutron spin polarization vector **P** is along the Z direction, and so is the external magnetic field **H** (guide field). Vertical and horizontal atomic spin **S** (magnetization) components give rise to non-spin-flip (NSF) and spin-flip (SF) scattering, respectively. The SF scattering amplitudes U^{+-}/U^{-+} are purely magnetic, whereas in the NSF scattering (U^{++}/U^{--}) there is an interference of magnetic and nuclear terms. The + and − signs describe neutron spins 'up' and 'down', respectively; b and p denote neutron's nuclear and magnetic scattering amplitudes, respectively.

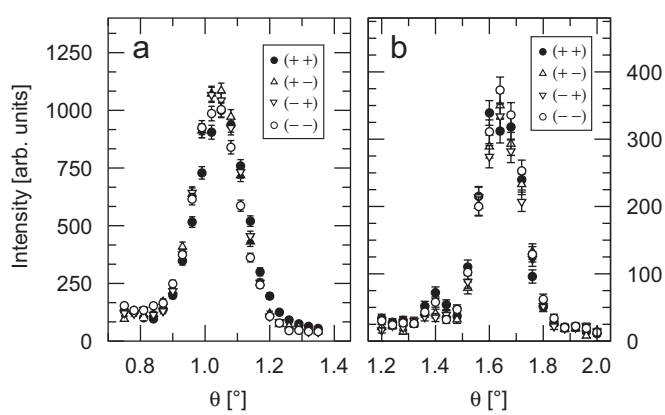

Fig. 3. Neutron polarization analysis of the first SL Bragg peak for: (a) EuS/YbSe (44 Å/20 Å) × 15 SL with the in-plane ⟨1 1 0⟩ axes @45° to the (vertical) polarization direction; (b) EuS/PbS (35 Å/4.5 Å) × 10 SL with the in-plane ⟨2 1 0⟩ axes @45° to the neutron polarization direction.

magnetic and the ratio of NSF to SF intensities from such samples can be expressed in the following form:

$$\frac{I_{\mathrm{NSF}}}{I_{\mathrm{SF}}} = \frac{x\sin^2(\phi_0+\Delta\phi)+(1-x)\cos^2(\phi_0+\Delta\phi)}{x\cos^2(\phi_0+\Delta\phi)+(1-x)\sin^2(\phi_0+\Delta\phi)}, \tag{1}$$

where $\phi = \phi_0 + \Delta\phi$, ϕ_0 describes the initial sample alignment and $\Delta\phi$ is the angle of the sample rotation about the normal to the surface.

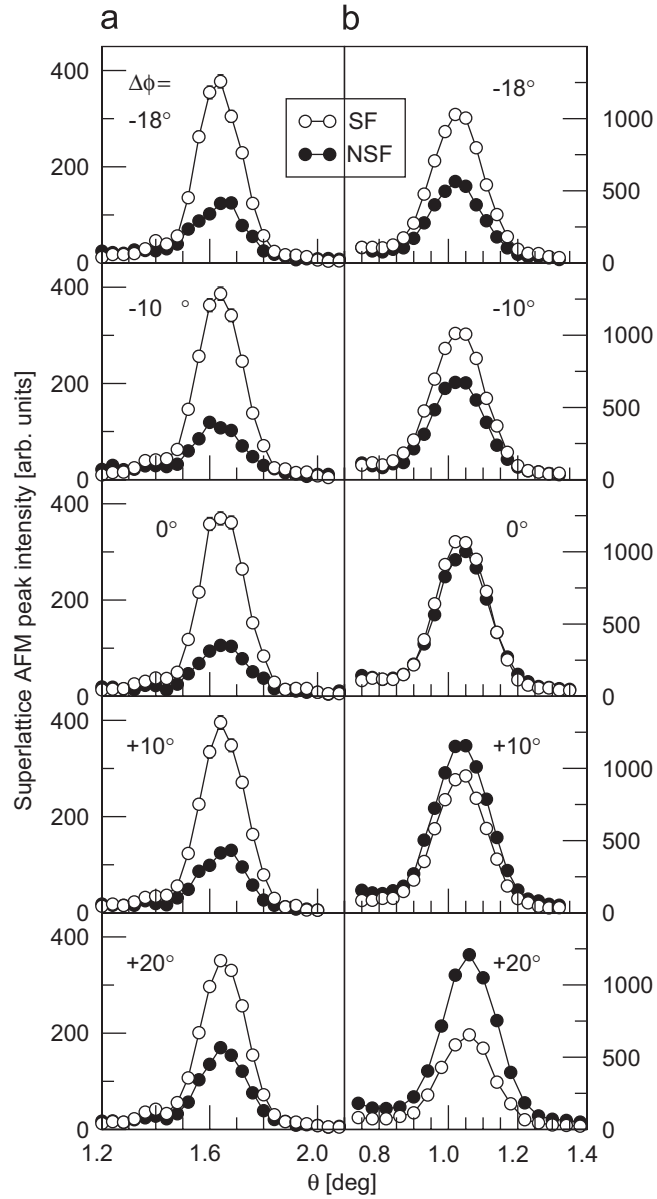

Fig. 4. The NSF and SF intensities of the AFM superlattice Bragg peaks for different sample orientations with respect to the neutron polarization direction: (a) EuS/PbS with [2 1 0] axis, and (b) EuS/YbSe with [0 1 0] axis horizontal (when $\Delta\phi = 0$).

The experimentally determined values of $I_{\mathrm{NSF}}/I_{\mathrm{SF}}$ and the fit of Eq. (1) to the data points are presented in Fig. 6. The values of the x and ϕ_0 parameters obtained from the fits are also shown in Fig. 6 for both samples. The broken line shows the calculated ratio of NSF to SF intensities in the case of a uniaxial sample (for which $x = 1$) with the same alignments with respect to the polarization direction.

3. Interlayer coupling in EuS/PbS SL's grown on (1 1 1)BaF_2

The results of neutron reflectivity measurements carried out on EuS/PbS superlattices grown on the (1 1 1)

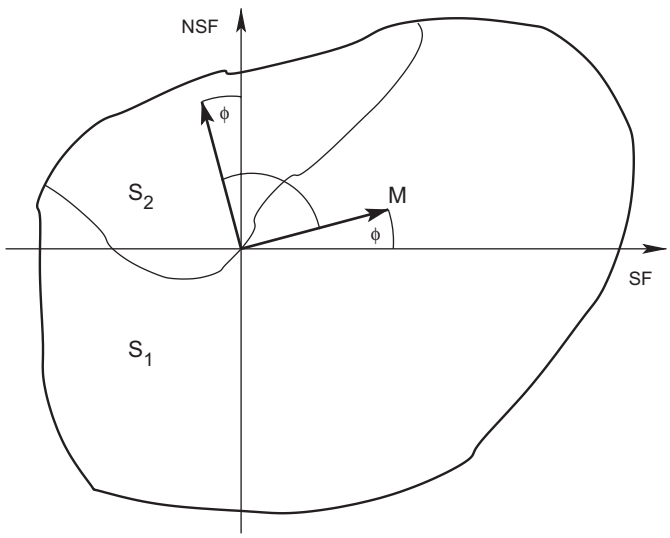

Fig. 5. A schematic view of the sample surface. The two areas labeled as S_1 and S_2 represent domains with magnetization vectors **M** along two perpendicular crystallographic axes.

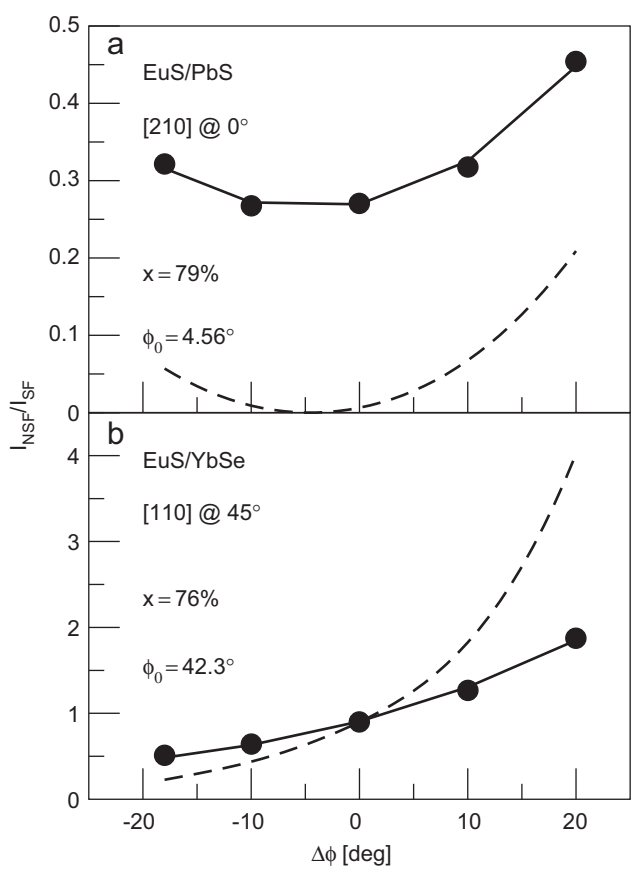

Fig. 6. NSF to SF intensity ratio as a function of the sample orientations with respect to the neutron polarization direction for two SL specimens of different composition. Solid lines represent the fits of Eq. (1) to the experimental data. Broken lines show NSF/SF ratios, calculated from Eq. (1), for uniaxial samples (i.e., for $x = 1$) with the same alignment. The fitted values of ϕ_0 reflect the actual initial sample misalignment.

plane of BaF_2 substrate differ significantly from the results of previously studied EuS/PbS SL's grown on (0 0 1)KCl substrates. In Fig. 7, typical neutron reflectivity patterns taken with unpolarized neutron beams are presented for the EuS/PbS superlattices with 65, 90, and 150 Å thick PbS spacer. The measurements performed above and below T_C show a considerable increase of the intensity of the first structural SL Bragg peak with decrease of the temperature. This clearly demonstrates the formation

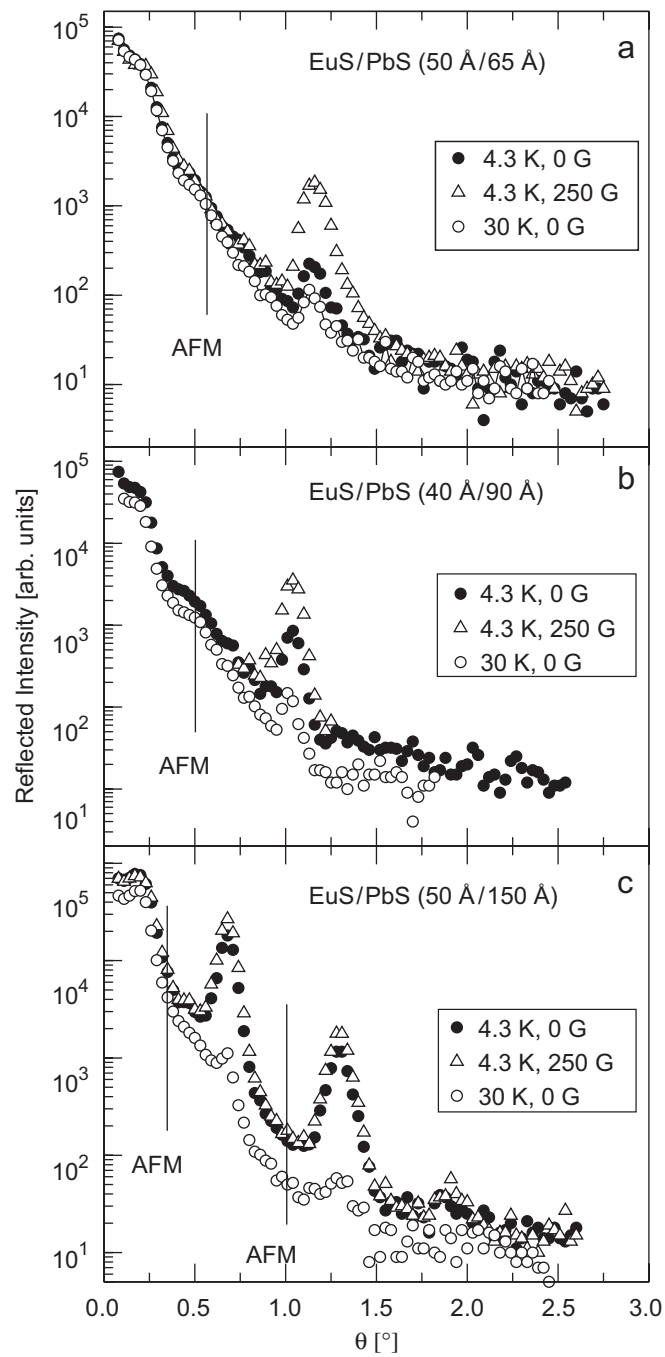

Fig. 7. Neutron reflectivity spectra for EuS/PbS superlattices grown on (1 1 1)BaF_2 substrates, measured below and above T_C in zero (filled circles and triangles, respectively) and in a saturating (open circles) magnetic field.

of the ferromagnetic (FM) interlayer correlations in the investigated SL. At the same time, no intensity increase can be detected at the AFM peak position (see the AFM positions in Fig. 7). Thus, there are even no traces of AFM interlayer correlations in these superlattices.

The existing theoretical model explaining the AFM IEC in EuS/PbS SL's grown on (0 0 1)KCl substrates [8] does not predict the FM IEC in SL's grown on (1 1 1)BaF_2; to the contrary it also predicts an AFM IEC although slightly weaker than in the case of SL's grown on KCl. Thus, the most important question concerns the origin of the FM alignment of the EuS layers in SL's grown on BaF_2. One cannot exclude that it might be caused by other than IEC factors, in particular, cooling the sample through T_C in a small magnetic field (e.g. earth field or other field present at the neutron reflectometer) might induce a parallel alignment of the magnetic domains along the field direction and mimic the effect of the ferromagnetic IEC.

In order to determine the in-plane magnetic domain orientation distribution, additional polarized neutron reflectivity measurements have been performed for a EuS/PbS (50 Å/150 Å) SL. An extensive set of data has been collected for the sample being rotated about the normal to its surface, with the rotation angles ranging from −180° to +180°. A very strong variation of spin dependent scattering intensities has been observed during the sample rotation as can be seen in Fig. 8.

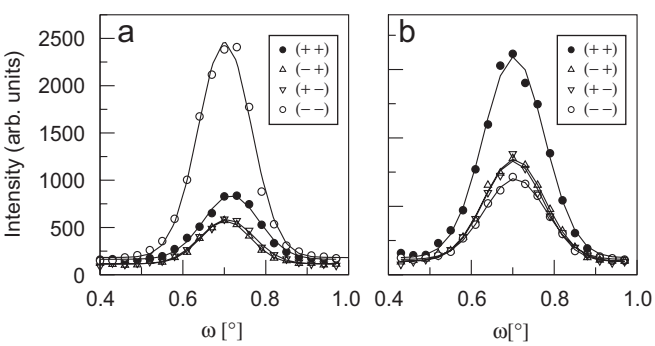

Fig. 8. The two examples of the dependence of the polarized neutron intensities on the angle of rotation of the sample; (a) and (b) the sample rotated by 60° and 150° from the starting position, respectively.

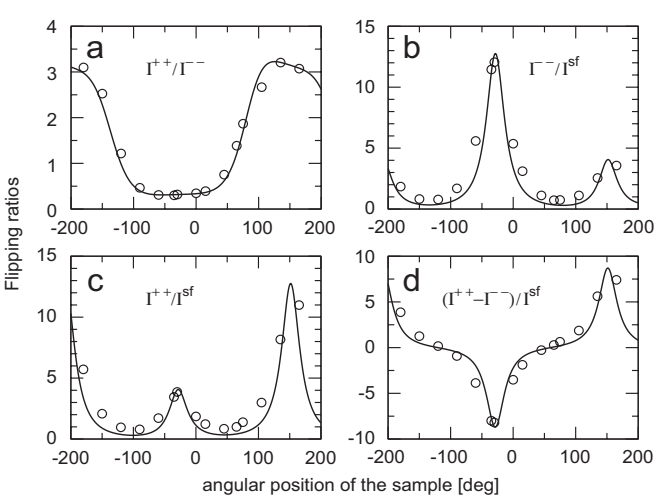

Fig. 9. The flipping ratios vs. angle of rotation for EuS/PbS (50 Å/150 Å) × 10 superlattice. The fit of a model domain orientation distribution is shown by a solid line.

The observed spin dependent intensities have been corrected for the polarizing efficiency of the supermirror polarizer and analyzer as well as for the flipping efficiences of the flippers. The details of the procedure can be found in Ref. [9]. Thus obtained corrected intensities were used for calculation of the several flipping ratios.

In Fig. 9(a–d) the obtained ratios of I^{++}/I^{--}, I^{++}/I^{sf}, I^{--}/I^{sf} and $(I^{++}-I^{--})/I^{sf}$ vs. angular position of the SL sample are presented together with the fit of the model of the magnetic domains distribution. I^{sf} is the average value of I^{-+} and I^{+-}.

3.1. Model of the magnetic domains distribution in ferromagnetically coupled EuS/PbS on (1 1 1)BaF_2

In order to explain the observed flipping ratios a model of the in-plane magnetic domain distribution has been developed in which the domains are oriented along the six, symmetry allowed, in-plane directions. In contrast to AFM coupled SL's where all the scattering intensities were purely magnetic in ferromagnetically correlated SL's, the non-spin flip SL Bragg peak intensities depend both on nuclear and magnetic SLD contrast. In Table 1, nuclear and magnetic scattering length densities (SLD) calculated from the nuclear scattering lengths and magnetic moments of the constituent superlattice materials EuS and PbS are presented.

The SLD contrast between layers thus equals to

$$\Delta\mathrm{SLD}_{\mathrm{nucl}} = (-0.0449 + i0.0237) \times 10^{-5}\,\text{Å}^{-2}, \quad (2)$$

$$\Delta\mathrm{SLD}_{\mathrm{magn}} = 0.3550 \times 10^{-5}\,\text{Å}^{-2}, \quad (3)$$

and for a polarized neutron beam the spin dependent intensities become

$$I^{++} \propto |\Delta\mathrm{SLD}_{\mathrm{nucl}} - \Delta\mathrm{SLD}_{\mathrm{magn}}\cos\phi|^2, \quad (4)$$

$$I^{--} \propto |\Delta\mathrm{SLD}_{\mathrm{nucl}} + \Delta\mathrm{SLD}_{\mathrm{magn}}\cos\phi|^2, \quad (5)$$

$$I^{+-/-+} \propto |\Delta\mathrm{SLD}_{\mathrm{magn}}\sin\phi|^2. \quad (6)$$

In order to check if the calculated values of nuclear and magnetic SLD contrast describe correctly our samples, the spin dependent SL Bragg peak intensities were measured

Table 1
SLD's of EuS and PbS in 10^{-5} Å^{-2}

	Magn. SLD	Nucl. ReSLD	Nucl. ImSLD
EuS	0.3550	0.1894	0.0237
PbS	0.0	0.2343	0.0

with the sample in saturating magnetic field. For a saturated sample there is only a single magnetic domain oriented along the external field thus making an angle $\phi = 0$ with the neutron polarization direction. In such a case there should be no spin-flip scattering ($I^{+-} = I^{-+} = 0$). The NSF I^{++} and I^{--} intensities should be different due to the interference between magnetic and nuclear scattering. The calculated value of the I^{++}/I^{--} flipping ratio is then

$$\frac{I^{++}}{I^{--}} = \frac{|\Delta\mathrm{SLD}_{\mathrm{nucl}} - \Delta\mathrm{SLD}_{\mathrm{magn}}|^2}{|\Delta\mathrm{SLD}_{\mathrm{nucl}} + \Delta\mathrm{SLD}_{\mathrm{magn}}|^2} = 1.659. \tag{7}$$

The uncorrected data are presented in Fig. 10, the very small spin-flip intensities seen in the figure come from imperfect polarizer and flipper efficiencies. The splitting between I^{++} and I^{--} is clearly visible.

The corrected experimental value for the I^{++}/I^{--} flipping ratio equals 1.653 and is very close to the calculated one. This excellent agreement allows us to use the values of nuclear SLD's shown in Table 1 in the subsequent calculations. The magnetic SLD for an unsaturated, multidomain, sample may differ significantly; this issue will be discussed later.

Due to the threefold symmetry in the (1 1 1) plane we assume the existence of the six easy directions for the in-plane magnetic domains, each easy direction making an angle ϕ_i with the (vertical) neutron polarization direction. The fractional population of each domain is x_i where $\sum_{i=1}^{6} x_i = 1$. At this stage the actual crystallographic orientation of these easy directions was not specified yet.

The contribution of each domain to the spin dependent reflected intensities is then:

$$I_i^{++} \propto x_i|\Delta\mathrm{SLD}_{\mathrm{nucl}} - \Delta\mathrm{SLD}_{\mathrm{magn}}\cos\phi_i|^2, \tag{8}$$

$$I_i^{--} \propto x_i|\Delta\mathrm{SLD}_{\mathrm{nucl}} + \Delta\mathrm{SLD}_{\mathrm{magn}}\cos\phi_i|^2, \tag{9}$$

$$I_i^{+-/-+} \propto x_i|\Delta\mathrm{SLD}_{\mathrm{magn}}\sin\phi_i|^2. \tag{10}$$

In order to obtain the experimentally measured spin dependent intensities we add the contributions from all the domain families:

$$I^{++} = \sum_i I_i^{++} \quad I^{--} = \sum_i I_i^{--},$$
$$I^{+-} = \sum_i I_i^{+-} \quad I^{-+} = \sum_i I_i^{-+}. \tag{11}$$

We assume that the angle $\phi_i = \phi_0 + i \times 60^\circ + \alpha$, where ϕ_0 is an initial sample orientation and α is an angle of rotation of the sample about the scattering vector **Q**.

The calculated, from the model, flipping ratios vs. angle of rotation α were fitted to the experimentally determined ones; x_i, ϕ_0, and $\Delta\mathrm{SLD}_{\mathrm{magn}}$ were treated as adjustable parameters. The crytallographic sample orientation corresponding to angle $\alpha = 0$ was determined by X-ray Laue photography. For this sample position the in-plane directions $[\bar{2}\,1\,1]$ and $[0\,1\,\bar{1}]$ were along vertical and horizontal axes, respectively. The value of ϕ_0 obtained from the fit was 150.5° measured clockwise from the vertical axis. Thus, the six easy directions (as obtained from the fit) coincide with the six in-plane $\langle 0\,1\,1\rangle$ directions. The obtained x_i values and the directions of the domain magnetizations are shown in the Fig. 11.

The value of $\Delta\mathrm{SLD}_{\mathrm{magn}}$ obtained from the fit equals $0.070 \times 10^{-5}\,\text{Å}^{-2}$ and is significantly lower than the

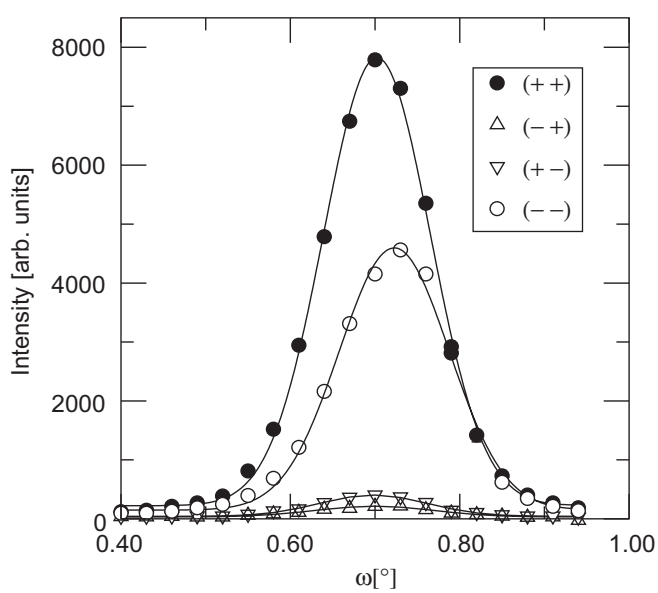

Fig. 10. The first SL peak measured in a saturating magnetic field. Clearly seen is the absence of SF scattering; only some residual intensity is visible due to the imperfect neutron beam polarizations and flipper efficiences. NSF intensities I^{++} and I^{--} show the splitting due to the interference between the nuclear and magnetic scattering.

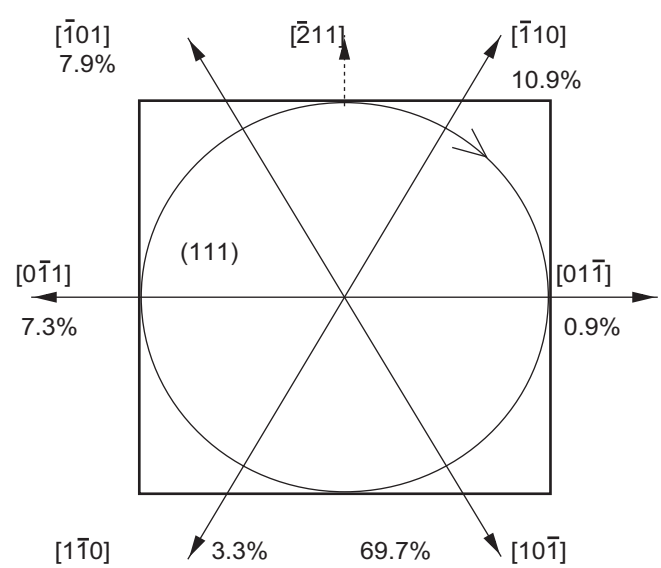

Fig. 11. The (1 1 1) plane of the superlattice with the six symmetry allowed $\langle 0\,1\,1\rangle$ directions. At the each $\langle 0\,1\,1\rangle$ direction the percentage of the magnetic domains oriented along this direction is given, The "zero-angle" sample position was with the $[\bar{2}\,1\,1]$-direction vertical. The sample was rotated about the normal to the surface [1 1 1] direction.

theoretically calculated one (0.355×10^{-5} Å^{-2} for the fully magnetized EuS layer). This discrepancy may be resolved if one assumes that the sizes of all domains in each family are comparable or smaller than the lateral coherence length (about 100 μm). In such a case, the EuS layer magnetization averaged over lateral coherence length, as 'seen' by the instrument, is lower than the saturation (single domain) magnetization due to a partial cancellation of the smaller domains of opposite orientation. Unfortunately, there is no information available about the sizes of the domains in EuS/PbS systems.

The most important conclusion from these data is that the domain direction distribution, although not equal for all symmetry-allowed directions, is definitely not uniaxial. Thus, a possible spurious lineup of domains during cooling in an external magnetic field should be excluded and FM IEC between EuS layers is the most probable cause of the observed ferromagnetic interlayer correlation. A reason for the departure from the fully symmetric domain direction distribution is not fully understood yet but a similar non-uniform domain distribution has also been observed in the AFM coupled EuS/PbS SL's on (0 0 1)KCl discussed earlier.

4. Conclusions

By applying the technique of neutron polarization analysis, together with the rotation of the samples about the scattering vector, it was possible to observe the magnetic scattering contributions from domain states in ferro- and antiferromagnetically coupled magnetic semiconductor superlattices. In all the systems under investigation the directional distribution of magnetic domains showed a pronounced departure from the in-plane crystallographic symmetry. There are several factors that may introduce such in-plane symmetry breaking, among them surface reconstruction with the different than in bulk material symmetry, asymmetric strain caused by lattice mismatch of the constituent materials or substrate miscut in a particular crystallographic direction. The full understanding of the observed phenomenon is not available yet. Nevertheless, neutron polarization analysis proved a very powerful tool for investigating such surface structure related effects.

Acknowledgment

Work supported by NSF DMR-0509478 Grant.

References

[1] C.J.P. Smits, et al., Phys. Rev. B 69 (2004) 224418.
[2] H. Kepa, et al., J. Alloys Compd. 401 (2005) 238.
[3] H. Ohno, J. Magn. Mag. Mater. 200 (1999) 110 and references therein.
[4] H. Kepa, et al., Phys. Rev. B 64 (2001) 121302.
[5] W.T. Lee, et al., Phys. Rev. B 65 (2002) 224417.
[6] S. Langridge, et al., Rev. Lett. 85 (2000) 4964.
[7] R.M. Moon, T. Riste, W.C. Koehler, Phys. Rev. 181 (1969) 920.
[8] J. Blinowski, P. Kacman, Phys. Rev. B 64 (2001) 045302.
[9] C.F. Majkrzak, Physica B 221 (1996) 356.