Birefringent Bragg Diffraction of Evanescent Neutron States in Magnetic Films

R. Günther,[1,2] W. Donner,[1] B. P. Toperverg,[3] and H. Dosch[1,4]
[1]Fachbereich Physik, Bergische Universität Wuppertal, D-42285 Wuppertal, Germany
[2]Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex 9, France
[3]Petersburg Nuclear Physics Institute, Gatchina, St. Petersburg 188350, Russia
[4]Max-Planck-Institut für Metallforschung, D-70569 Stuttgart, Germany
(Received 24 December 1997; revised manuscript received 12 March 1998)

When monochromatic neutrons impinge onto a surface below the critical angle for total external reflection, evanescent (tunneling) neutron wave fields emerge in a thin film below the surface. We show by theory and experiment on an Fe(001) film that this scheme leads to an unusual quadruplet structure of the evanescent Bragg diffraction when unpolarized neutrons tunnel into a ferromagnetic single crystal film. The observed Bragg splitting leads to a complete autoseparation of all neutron spin states and spin-flip processes without applying any spin analysis. We demonstrate that this surface phenomenon is most sensitive to perpendicular moments and indicate its potential for future applications. [S0031-9007(98)06451-5]

PACS numbers: 61.12.Ex, 75.70.Ak, 75.70.Cn

For many technical applications, as in data recording or spin valve devices [1], the understanding of the detailed magnetic structure across the film and the interfacial magnetic roughness are of crucial importance; consequently, many sophisticated scattering techniques have been devised to unravel the interfacial spin structure, such as resonant magnetic x-ray scattering [2,3] or spin polarized low energy electron diffraction [4]. Polarized neutrons [5] have successfully been employed to disclose the depth profile $M(z)$ of the magnetic moments in thin films, across buried interfaces, and in tailored magnetic multilayers [6]. This information is accessible via the specular ("mirror") reflection of polarized neutrons combined with a full analysis of the neutron spin after reflection. Since any magnetic neutron scattering process is governed by the term $\mathbf{e} \times [\mathbf{m} \times \mathbf{e}] = \mathbf{m} - \mathbf{e}(\mathbf{em}) = \mathbf{m}_\perp$ (where $\mathbf{e} = \mathbf{Q}/Q$ is the direction of the neutron momentum transfer $\mathbf{Q}$ and $\mathbf{m} = \mathbf{M}/M$ the direction of the magnetization $\mathbf{M}$), magnetic moments and magnetic fluctuations normal to the film are not accessible to neutrons along the specular rod ($\mathbf{Q} = \mathbf{Q_z}$), but require a noticeable in-plane neutron momentum transfer.

In this Letter we present an experimental study of glancing angle neutron Bragg scattering signals [7]. We find a novel multiplet structure which originates from birefringent evanescent neutron states as they emerge in magnetic materials when unpolarized neutrons are reflected from the magnetic interface. We discuss the origin of this neutron diffraction phenomenon and show by theory and experiment that these scattering signals give helpful information on the magnetic structure of the film and on the interfacial magnetic roughness.

Consider an unpolarized monochromatic neutron with wave vector $k = 2\pi/\lambda$ impinging onto the surface of a ferromagnetic film which has a thickness d and an average magnetization $\overline{\mathbf{B}}$ [Fig. 1(a)]. In the vacuum half space the neutron momentum normal to the film is q_0 [Fig. 1(a)]; within the magnetic film it reads—due to the Zeeman effect associated with a $s = 1/2$ particle—$q_\pm = \sqrt{q_0^2 - q_{c\pm}^2}$, with $\hbar^2 q_{c\pm}^2 = 2m(\overline{V}_N \pm \mu\overline{B})$ [8]. This phenomenon is known as magneto-optical birefringence [see Fig. 1(a)]. *Birefringent neutron propagation* in magnetic materials was first reported by Schneider and Shull [9]; in this Letter, we present and discuss *evanescent neutron birefringence* which occurs in the regime of total external reflection, i.e., when the neutron encounters a mirrorlike surface under an incident angle α_i less than the associated critical angle $\alpha_{c\pm}$ (given by $\sin\alpha_{c\pm} = q_{c\pm}/k$). Then, $q_\pm$ has a strong imaginary ("evanescent") component and a totally reflected beam occurs in the vacuum half space as well as two birefringent evanescent neutron wave fields inside the magnetic film which are proportional to the birefringent transmission coefficients $t_\pm = \{2q_0(q_\pm + q_s)\}/\Delta_\pm$ [10]. For the understanding of the experimental key results described below it is important to note only that the quantity $t_\pm$ exhibit distinct maxima for $\alpha_i = \alpha_{c\pm}$ [Fig. 1(a) inset] which leads to interesting new phenomena in the Bragg diffraction of such birefringent evanescent neutron wave fields from the lateral (nuclear and magnetic) crystal structure within the film. This will be discussed now.

The associated glancing angle diffraction geometry is shown in Fig. 1(b) [7]. Within the distorted wave Born approximation (DWBA) the Bragg diffraction amplitude is given by $f(k^f, k^i) \propto \langle\psi^f|\tilde{\mathcal{H}}|\psi^i\rangle$, where $(|\psi^i(z)\rangle)$ and $(|\psi^f(z)\rangle)$ are the initial and final neutron waves, respectively. Notice now that the incident neutron beam experiences the birefringent action of the transmission coefficient $t_\pm^i$ and, by reciprocity, also does the refracted beam upon entering into vacuum half space (thereby experiencing $t_\pm^f$). Since $t_\pm^i$ and $t_\pm^f$ induce a pronounced spin-dependent focusing of the incident and diffracted neutron wave, respectively [as illustrated in Fig. 1(a) for $t_\pm^i$], one would expect that the Bragg diffracted evanescent

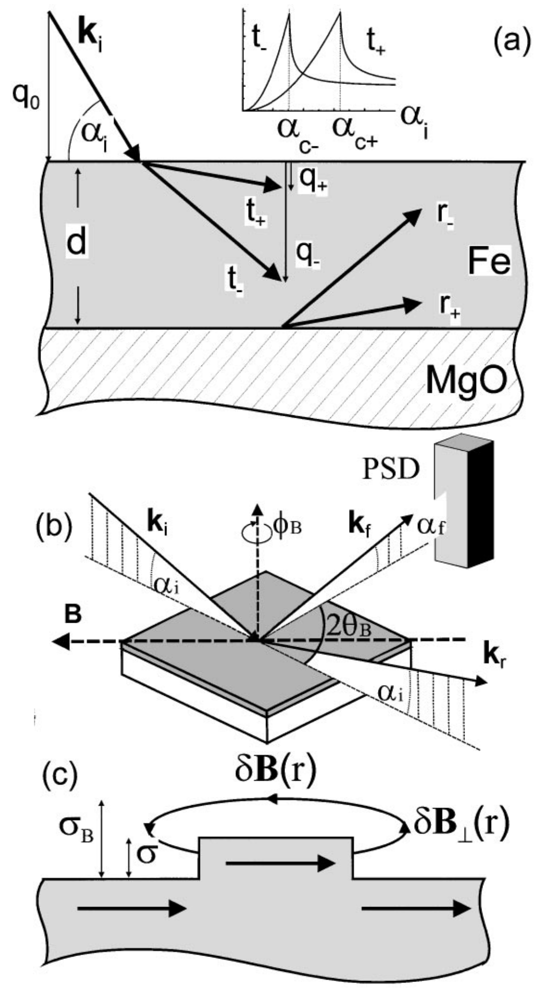

FIG. 1. Neutron wave field inside a magnetic film: the incident perpendicular momentum q splits into two birefringent evanescent states with the same in-plane component; both are reflected at the substrate. Inset: Eigenvalues of the transmission operator $\hat{t}$: both t_+ and t_- exhibit sharp maxima leading to a focusing of the diffracted intensity. (b) Sketch of the glancing angle Bragg diffraction scattering geometry with the in-plane diffraction angle $2\theta_B$; an external magnetic field **B** can be applied along any in-plane direction defined by the angle Φ_B. A position sensitive detector placed perpendicular to the sample surface allows for the collection of an α_f profile. (c) Sketch of the local magnetic stray field components occurring at a structural asperity.

neutrons should show a spatial quadruplet structure associated with all possible spin combinations $(++, --, +-, -+)$. Accordingly, the Bragg structure factor S should exhibit four separate maxima given by the conditions $\alpha_{i,f} = \alpha_{c\pm}$ [see Fig. 1(b)]. Two of them (S_{++} and S_{--}) are predicted to occur at $\alpha_i = \alpha_f = \alpha_{c+}$ and $\alpha_i = \alpha_f = \alpha_{c-}$, respectively, and originate from the independent Bragg diffraction of the two birefringent components of the evanescent neutron wave. The other two (S_{+-} and S_{-+}) should emerge at $\alpha_i = \alpha_{c+}, \alpha_f = \alpha_{c-}$ and $\alpha_i = a_{c-}, \alpha_f = \alpha_{c+}$ and result from spin-flip Bragg diffraction which transforms one spin component of the birefringent incoming wave into the other component of the outgoing wave.

In order to subject these intuitive ideas to a critical test we have performed a glancing angle neutron diffraction experiment using the EVA reflectodiffractometer at the high flux reactor of the Institute Laue-Langevin (ILL) [11]. The sample used for this study was a Fe(001) single crystal film with thickness $d = 3000$ Å and a mirrorlike surface grown epitaxially on MgO(001). The in-plane mosaicity ($\omega = 0.05°$) and the surface roughness ($\sigma = 4$ Å) of the Fe film have been measured by x rays. The Fe surface was terminated by a smooth oxide layer with a thickness of 26 Å as determined by x-ray reflectivity. For the grazing angle neutron diffraction experiment at EVA we used an unpolarized monochromatic neutron beam ($\lambda = 2.75$ Å) with a high collimation of $\Delta\alpha_i = 0.5$ mrad normal to the film plane (for magnetically saturated Fe the critical angles are $\alpha_{c\pm} = 0.15°, 0.32°$). The Fe film was exposed to an external magnetic field $B = 360$ mT along various in-plane directions (the saturation field along the hard magnetization axis is 80 mT as determined by Kerr-effect measurements). We recorded several in-plane Bragg diffraction profiles as a function of the incident angle α_i by a linear position sensitive detector which provided for each setting of α_i the associated exit angle (α_f) distributions [see Fig. 1(b)]. For the Fe(110) evanescent in-plane Bragg reflection a typical result of our observations is shown in Fig. 2(a) as a two-dimensional isointensity map vs α_i and α_f. In the inset in Fig. 3(a) the intensity along the marked line in Fig. 2(a) is extracted. Interestingly, the observed (110) Bragg intensity distribution discloses indeed a novel multiplet structure which we refer to the birefringent Bragg diffraction of neutron tunneling states. It leads to a complete spatial separation of all neutron spin states and spin-flip processes without using a spin polarizer or spin analyzer. We note here that these observations are rather different from another interesting spin-splitting effect in the specular beam described by Felcher *et al.* [12].

A straightforward but somewhat lengthy calculation confirms indeed this intuitive idea [13]: Assuming more generally an arbitrary spin polarization $\mathbf{P}_0$ of the incident beam and no polarization analysis after the diffraction process one obtains a structure factor $S(\mathbf{q})$ which can be cast into the form

$$S(\mathbf{q}) \propto \{(S_{++} + S_{+-})(1 + P_0) + (S_{--} + S_{-+}) \times (1 - P_0)\}, \tag{1}$$

with the non-spin-flip ("diagonal") structure factors $S_{\pm\pm} = |b_N \pm b_M^{\perp}|^2 \tilde{S}_{\pm\pm}$ given by the nuclear and magnetic scattering and the spin flip ("nondiagonal") structure factors $S_{\pm\mp} = b_M^{\perp} b_M^{\parallel} \tilde{S}_{\pm\mp}$ governed by magnetic scattering only. Since $b_M^{\perp} = b_M \mathcal{F}(\tau)[1 - (\mathbf{em})^2]$ and $b_M^{\parallel} = b_M \mathcal{F}(\boldsymbol{\tau})(\mathbf{em})^2$ this spin-flip scattering is forbidden for $\boldsymbol{\tau} \parallel \mathbf{m}$ and $\boldsymbol{\tau} \perp \mathbf{m}$ [14].

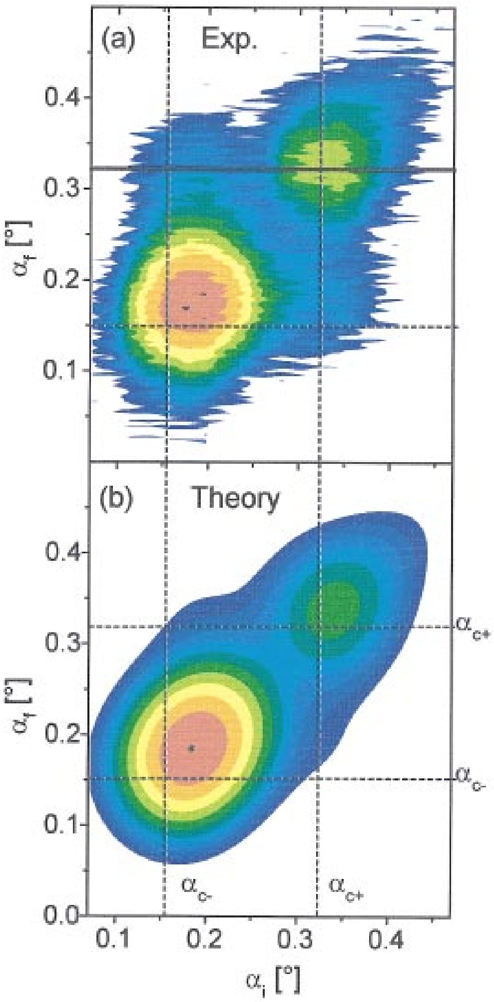

FIG. 2(color). α_i-α_f contour plot of the Fe(110) in-plane Bragg intensity; both graphs are on the same logarithmic scale. Dashed lines indicate α_{c-} and α_{c+} of iron; their crosspoints mark the angular positions of S_{++}, S_{--}, $S_{\mp}$, and $S_{\pm}$. (a) Experimental result using unpolarized neutrons. An external magnetic field **B** is applied along the in-plane [100] direction. (b) Theoretical simulation based on Eqs. (1) and (2).

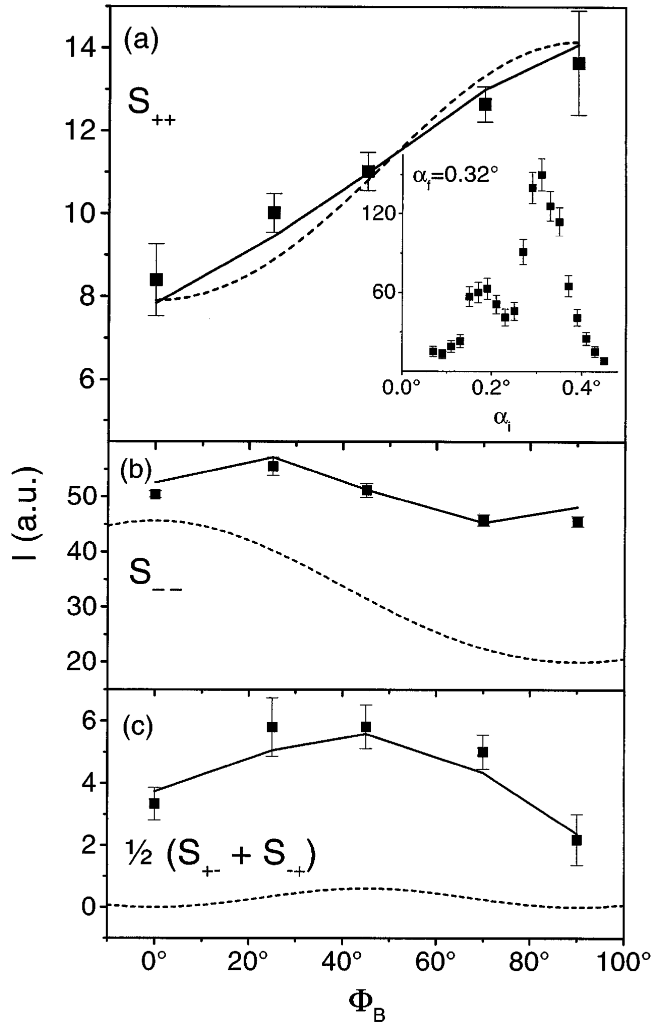

FIG. 3. S_{++} (a), S_{--} (b), and $\frac{1}{2}(S_{\pm} + S_{\mp})$ (c) as a function of the magnetic field direction **b**; full symbols are the observed values, the dashed line is calculated according to Eq. (1) (homogeneous magnetization), and the full line connects calculated values which take local stray fields at the interface into account [Eq. (3)]. The inset in (a) shows the intensity along the thick line in Fig. 2(a).

The key features in (1) are the coefficients ($\{\mu, \nu\} = \{+, -\}$)

$$\tilde{S}_{\mu\nu} = |t_\mu^f t_\nu^i g_{\mu\nu}^{tt} + r_\mu^f r_\nu^i g_{\mu\nu}^{rr} + r_\mu^f t_\nu^i g_{\mu\nu}^{rt} + t_\mu^f r_\nu^i g_{\mu\nu}^{tr}|^2, \tag{2}$$

which contain the birefringent transmission values ($t_{\pm}^{i,f}$) of the magnetic film (for further definitions, see [15]).

A numerical simulation of the intensity distribution based on expressions (1) and (2) is shown in Fig. 2(b). The positions and the widths of the two pronounced maxima are in excellent agreement with the experiment and confirm the unusual underlying beam focusing mechanism evoked by the twofold action of the ferromagnetic Fe-vacuum interface. In Fig. 2(a) we observe rather strong spin-flip contributions ($S_{\pm}$ and $S_{\mp}$) to the evanescent Bragg diffraction which are not covered by theory. In the above model [Eqs. (1) and (2)] a spin flip is evoked only by the magnetic Bragg scattering whenever the active in-plane reciprocal lattice vector $\boldsymbol{\tau}$ makes an angle $\Phi \neq 0°, 90°$ with the magnetization direction **m**. For a more quantitative understanding of the origin of the additionally observed spin-flip processes we investigated in a further experiment the intensity variation of the four Bragg maxima as a function of the in-plane rotation (Φ_B) of the external magnetic field **B**. For a quantitative presentation the observed intensity variations of S_{++}, S_{--}, and $\frac{1}{2}(S_{\pm} + S_{\mp})$ as integrated over the observed half-width are summarized in Fig. 3 together with the corresponding theoretical integrated intensities according to Eqs. (1) and (2) (dashed lines). While the general trend is well described by the scattering theory, we find that the measured spin-flip scattering level is significantly enhanced as compared to theory. As we will see in what follows these additional spin-flip processes are evoked by the "magnetic roughness" within the thin oxide layer.

In magnetic fields with magnetically rough or chemically altered interfaces spin-flip diffraction can result from the interference between the nuclear scattering and the neutron interaction with the microscopic stray field $\delta\mathbf{B}(\mathbf{r})$ as created by magnetic roughness or by "loose spins" at the surface and the film-substrate interface, as shown schematically in Fig. 1(c). The field fluctuations $\delta\mathbf{B}(\mathbf{r})$ are distributed with the correlation function $\mathcal{G}^{\alpha\beta}(\rho,z) = \langle \delta B^{\alpha}(\rho) B^{\beta}(0)\rangle$ within a range given by an unknown thickness σ_B [see Fig. 1(c)]. Assuming for simplicity axial symmetry of the magnetic stray field correlations, the correlator $\mathcal{G}^{\alpha\beta}$ can be decomposed into $\mathcal{G}^{\alpha\beta}(\rho,z) = G_0\{b^{\alpha}b^{\beta}[1 - g_{\perp}(\rho,z)] + (\delta^{\alpha\beta} - b^{\alpha}b^{\beta})g_{\perp}(\rho,z)\}$, where $g_{\perp}$ measures the density of field components perpendicular to the average in-plane magnetization which flip the spin of the impinging neutron. The associated scattering cross section can again be calculated within the DWBA, for unpolarized neutrons ($\mathbf{P}_0 = 0$) the final result can be cast into the simple form [13]

$$S_r(\mathbf{q}) \propto b_N^2 |a^{fi}|^2 \frac{\sigma_B}{d} [(1 - g_{\perp})(\tilde{S}_{++} + \tilde{S}_{--}) + g_{\perp}(\tilde{S}_{+-} + \tilde{S}_{-+})], \quad (3)$$

which contains σ_B and $g_{\perp}$ are free fitting parameters [the quantity $a^{fi} = (q_{f+} - q_{f-} + q_{i+} - q_{i-})d$ is a phase which depends on the glancing incident and exit angle]. The full line in Fig. 3 shows the best fit using (3) implying $\sigma_B = 51 \pm 6$ Å and $g_{\perp} = 0.4 \pm 0.1$.

We associate these two experimental parameters to the neutron spin-flipping power of the oxide cap which acts as a magnetically rough interface. Interestingly we find that the magnetically active thickness is within the experimental error twice as thick as the oxide cap, $\sigma_B \approx 2\sigma$. Apparently, the local depolarization fields $\delta\mathbf{B}(\mathbf{r})$ extend significantly out into the vacuum half space as shown schematically in Fig. 1(c). This observation is in seeming contrast to a recent diffuse x-ray resonant magnetic scattering study of Co/Cu interfaces [3] which appear to be magnetically less rough than structurally; however, in the latter case, it has been suggested that the local out-of-plane moments do not contribute to the diffuse x-ray signals. The evanescent neutron technique presented here on the other hand is most sensitive to such moments "sticking out"; the observed value $g_{\perp} = 0.4$ implies in fact that 40% of the moments within the surface layer have components perpendicular to the mean magnetization.

In summary we have presented a novel quadruplet splitting of the in-plane Bragg law which is observed for unpolarized neutrons which undergo birefringent tunneling into a ferromagnetic single crystal Fe(001) film. It is shown by a detailed analysis of the spin-flip Bragg channels that the local fields within the layer sticking out of the surface extend significantly into the vacuum half space. Since such local stray fields are most important in magnetic thin film devices [1], the presented neutron technique may be used to investigate perpendicular magnetic roughness in thin films [16] and in superlattices of current interest. Such experiments are currently in preparation.

We are grateful to the ILL for continuous support and hospitality. We thank O. Schärpf for helpful advice and H. Zabel for allowing us to perform the Kerr-effect measurements. This work is supported by the BMBF under Grant No. 03-DO4WUP-2.

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[15] $g_{\mu\nu}^{tt} = \{\exp(i\phi_+) - 1\}/\phi_+$, $g_{\mu\nu}^{rr} = g_{\mu\nu}^{tt}\exp(-i\phi_+)$, $g_{\mu\nu}^{tr} = \{\exp(i\phi_-) - 1\}/\phi_-$, $g_{\mu\nu}^{rt} = g_{\mu\nu}^{tr}\exp(-i\phi_-)$, and $r_{\pm} = 2q_0(q_{\pm} - q_c)\exp(2i\phi_{\pm}/\Delta_{\pm})$.
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