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Spin-polarized electron energy loss spectroscopy: a method to measure magnon energies

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Abstract

Spin-polarized electron energy loss spectroscopy is demonstrated to be an effective and unique tool to determine spin-wave dispersion curves of surfaces and ultrathin films over the whole Brillouin zone. The spin-wave dispersion curve of 8 monolayer Co on Cu(0 0 1) along the $\langle 1\,1\,0 \rangle$-direction has the shape of a thin film spin dispersion curve with zero slope at the surface Brillouin zone. The cross-section of magnon excitation depends strongly on the energy E_0 of the incident electrons and has a maximum around $E_0 = 7$ eV.

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1. Introduction

The properties of long wavelength spin waves at surfaces and in thin films have been investigated very extensively in the past by ferromagnetic resonance (FMR) and Brillouin light scattering (BLS) or optical time domain methods [1]. The velocity mismatch between electro-magnetic radiation and spin waves limits these methods to the investigation of long wavelength spin waves with wave vectors smaller than about 10^{-2} Å^{-1}. With magnetic neutron scattering spin waves within the whole Brillouin zone can be observed. However, the interaction of neutrons with spin waves lacks any surface sensitivity and is so weak, that investigations of spin waves at surfaces or in single thin films are practically impossible [2]. Therefore the region of high energy and high wave vector spin waves is currently not explored in thin films and at surfaces.

Besides these collective spin wave excitations “single particle” Stoner excitations—a coupled pair of an electron above the Fermi energy E_F and a hole below E_F with opposite spin—can be excited very efficiently by electron scattering. Their properties have been investigated extensively during the last 20 years by spin polarized electron energy loss spectroscopy (SPEELS) [3–6]. However, no evidence for spin waves was found in the experimental SPEEL spectra until recently a spin-wave signature has been observed by SPEELS in 5 ML Fe on W(1 1 0) [7]. Early theories predicted, that spin waves should be observable by SPEELS, but no reliable estimate of the cross section could be made [8,9]. Only recently Plihal et al. [7] showed, that the spin waves cross section can be comparable to the cross section of Stoner excitations or even larger and therefore spin waves should be easily observable [10]. In this paper we show, that SPEELS can be used as a method to explore the high energy and high wave vector spin wave region of thin films.

2. Experiment

A new high resolution electron energy loss spectrometer has been used for the measurements presented in

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this paper, which is described in detail in Ref. [11]. A (longitudinal) spin-polarized electron beam is generated by illuminating a GaAs photo-cathode with circularly polarized light from a diode laser operating at 830 nm. Depending on the helicity of the light the spin polarization of the electron beam is either parallel or anti-parallel to the propagation direction of the photo-electron [12]. We used unstrained GaAs cathodes with a degree of spin-polarization of $P = 0.28 \pm 0.04$ (for the measurements of Fe/W(1 1 0)) as well as strained GaAs cathodes with $P = 0.79 \pm 0.09$ (for Co/Cu(1 0 0)) [13]. The e-beam passes a 90°-deflector and a 180°-deflector, so that a transversely spin polarized e-beam results. The scattering plane is perpendicular to the spin orientation of the incident electrons. A 146°-deflector analyzes the electrons scattered from the surface with respect to their energy. The total energy resolution in the experiments is about 40 meV (full-width half-maximum, FWHM). The scattering angle between incident and scattered beam is kept fixed at 90°. By rotating the crystal surface normal with respect to the incident angle θ, the wave vector transfer parallel to the surface $\Delta K = k_f \cos\theta - k_i \sin\theta$ can be changed. k_i, k_f are the magnitudes of the wave vectors of the incident and scattered electron, respectively.

Co was deposited by molecular beam epitaxy (MBE) onto Cu(0 0 1) at 300 K, forming a pseudomorphic FCC structure [14]. The thickness of the Co film has been calibrated by medium energy electron diffraction (MEED). For the data shown in this paper the Co film was eight monolayers (ML) thick. The Co film has been annealed at 450 K for 5 min to produce a smoother surface. For a thickness larger than 4 ML these films are stable against pin-hole formation and inter-diffusion at that temperature [15].

Fe films with a thickness of approximately 5 ML were grown on a W(1 1 0) surface by MBE at room temperature. While the first ML of Fe grows pseudomorphic on W(1 1 0), the large lattice mismatch between Fe and W leads to a relaxation of the film by the formation of a dislocation network [16]. In difference to bulk Fe, the easy axis of magnetization for Fe on W(1 1 0) is along the ⟨1 1 0⟩-direction in the surface plane [17].

The films were magnetized along their easy axis of magnetization after preparation. For the SPEELS measurements the scattering plane was chosen perpendicular to the magnetization. SPEEL intensity spectra for the polarization of the incident beam anti-parallel ($I'_\uparrow$) and parallel ($I'_\downarrow$) to the magnetization were recorded, i.e. for $I'_\uparrow$ ($I'_\downarrow$) the incident electron has majority (minority) character with respect to the sample. The measured intensities were corrected for the incomplete polarization P: $I_{\uparrow(\downarrow)} = (I'_{\uparrow(\downarrow)}(P+1) + I'_{\downarrow(\uparrow)}(P-1))/(2P)$. The asymmetry A is defined here as $A = (I_\downarrow - I_\uparrow)/(I_\downarrow + I_\uparrow)$.

3. Results and discussion

3.1. 8 ML Co on Cu(0 0 1)

Firstly, results from an 8 ML thick Co film on Cu(0 0 1) are presented. The SPEEL spectrum for $I_\downarrow$ (solid symbol) for an incident angle $\theta = 17.5°$ and an incident energy $E_0 = 6.7$ eV, corresponding to a wave vector transfer of $\Delta K = 0.87$ Å^{-1} in Fig. 1 shows a clear well-resolved peak at about 189 meV besides the diffuse elastic peak at 0 meV, while no such peak is observable in the $I_\uparrow$ spectrum (open symbols in Fig. 1). The large intensity difference between $I_\downarrow$ and $I_\uparrow$ at large energy losses is due to the Stoner excitation continuum [5,6]. The continuum intensity tends to decrease towards smaller energy losses, but no threshold can be determined. The dashed line in Fig. 2 gives a qualitative estimate of the Stoner background. The fact, that the spin wave peak is only observed in the $I_\downarrow$ spectrum is explained by the conservation of angular momentum: The excitation of a spin wave reduces the magnetization of the Co film. Therefore, the scattered electron must be a majority spin electron and the incident electron minority spin electron.

The full spin-wave dispersion curve can be determined from the SPEEL intensity spectra taken at different incident angle θ corresponding to a different wave vector transfer ΔK, as shown in Fig. 2. In the $I_\downarrow$ spectra (Fig. 1(b)) a well-defined peak emerges from the diffuse elastic

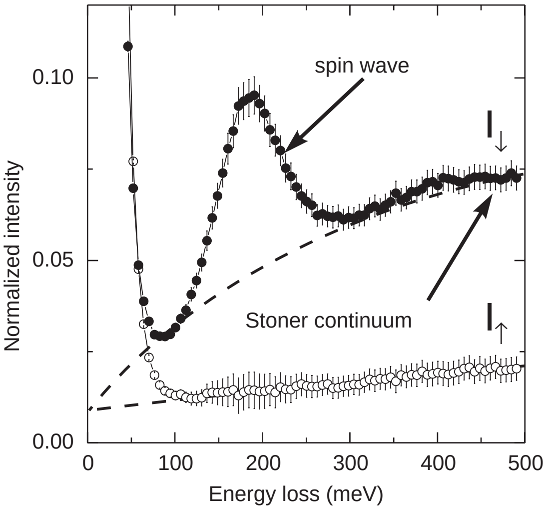

Fig. 1. SPEEL intensity spectrum, normalized to the average peak intensity $(I_\uparrow + I_\downarrow)/2$ at 0 meV energy loss, from 8 ML Co on Cu(0 0 1) along the ⟨1 1 0⟩-direction at $\Delta K = 0.87$ Å^{-1}, ○ (●) for incident electrons with majority (minority) character with respect to the sample. The vertical error bars indicate the error in $I_\uparrow$ and $I_\downarrow$ due to the uncertainty in the polarization P. The dashed line extrapolates qualitatively the Stoner background to 0 meV.

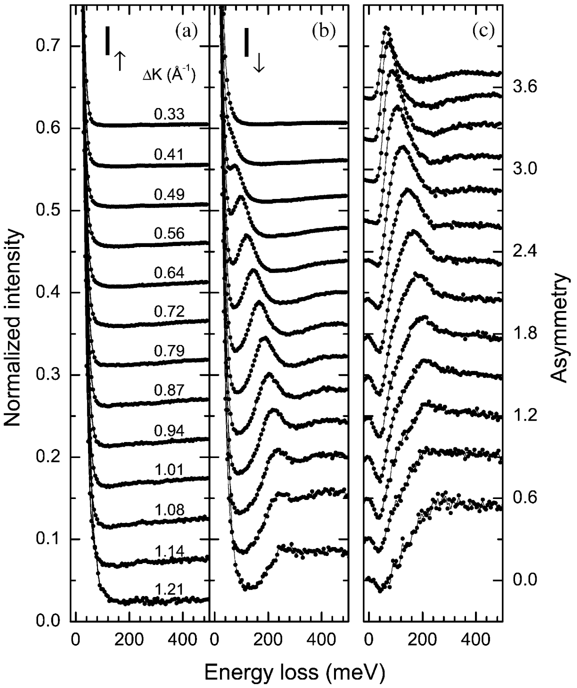

Fig. 2. Series of SPEEL intensity spectra from 8 ML Co on Cu(0 0 1) along the ⟨1 1 0⟩-direction for different ΔK, (a) for $I_\uparrow$ and (b) for $I_\downarrow$. (c) The asymmetry calculated from (a) and (b). The intensity for each ΔK is normalized to the average peak intensity at 0 meV energy loss.

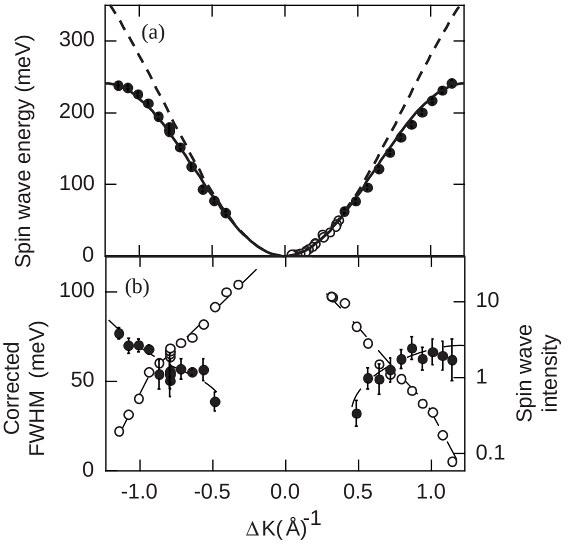

Fig. 3. (a) Spin-wave dispersion curve derived from the spin-wave peak position in the SPEEL spectra (●). For comparison the available neutron data for bulk FCC Co (8% Fe) are shown as well (○, Ref. [18]). The solid (dashed) line is a fit curve of a 2D- (3D-)Heisenberg model to the SPEELS data. (b) Width (left scale and ●), and relative intensity (right scale and ○) of the spin wave peak shown in Fig. 1. The dashed lines are guidelines to the eye only.

peak and moves towards higher energy losses with increasing ΔK while in $I_\uparrow$ (almost) no such peak is observable. The corresponding asymmetry in Fig. 2(c) has this peak structure as well, most clearly at low ΔK. At larger ΔK the asymmetry contribution due to Stoner excitations increases.

The data in Fig. 2(b) were analyzed by gaussian fits to the spin wave loss peak and the diffuse elastic peak at 0 meV. The background, which is mainly due to Stoner excitations, has been taken into account by a second order polynomial. The positions of the spin wave peak, derived from the fit is plotted in Fig. 3(a) as solid symbols. The solid line represents a fit curve to the data, which is based on a nearest neighbor 2D-Heisenberg model $H = -J\sum_{\langle i,j\rangle} S_iS_j$. The 2D-Heisenberg model results in a dispersion curve $E^{2D}(q) = 4JS(1-\cos(qa_0))$ for the ⟨1 1 0⟩-direction. J is the nearest neighbor exchange interaction energy, S (the expectation value of) the spin component parallel to the magnetization direction in one (primitive) unit cell, $q = |\Delta K|$ and $a_0 = 2.55$ Å the nearest neighbor distance. Although our 8 ML thick Co film cannot be described by a two-dimensional model, it can be shown that for sufficiently thick films the energy of the lowest spin-wave dispersion branch $E_1^{\rm film}(q)$ is approximately equal to twice the energy of the dispersion curve in a 2D-model, $E^{\rm film}(q) \approx 2E^{2D}(q)$. For the 8 ML film the deviation is already smaller than our experimental uncertainty. Using this model, we obtain $JS = 15.0$ meV. For comparison neutron data from bulk FCC Co (with 8% Fe to stabilize the FCC phase at room temperature) are included for the available wave-vector range [18]. These neutron data lie perfectly on the fit curve for the SPEELS data. However, the spin wave dispersion curve expected for the bulk differs significantly in shape, because the zone boundary in the ⟨1 1 0⟩-direction (K-point) is at a 1.5 times larger wave vector than the surface Brillouin zone boundary. The dashed curve is a fit curve to the neutron data based on the 3D-Heisenberg model: $E^{3D}(q) = 4JS(5-\cos(qa_0) - 4\cos(qa_0/2))$.

The mean free path for electrons with 6.7 eV energy is only 4–5 atomic layers in Co [19,20]. Therefore, SPEELS is a surface sensitive method, i.e. it is sensitive only to the spin-wave amplitude within the first few atomic layers from the surface of the sample. It follows that the spin-wave dispersion from a surface of a bulk ferromagnet measured by SPEELS may deviate from the dispersion measured with a bulk sensitive method like neutron scattering. At wave vectors above 0.8 Å^{-1} this deviation between the spin wave dispersion measured by SPEELS and the fit curve to the 3D-Heisenberg model is quite large, although the exchange interaction energy does not differ significantly for the two fit curves [18].

Actually, for thin films more than one spin wave dispersion curve is expected, because the spin waves are quantized in the direction perpendicular to the film surface. This should lead to additional standing spin waves branches in the spin-wave spectrum, as is observed in Brillouin light scattering for thick films and small wave vectors [21]. We do have no clear evidence for these standing spin waves in our SPEEL spectra. The spin wave peak is slightly asymmetric with some additional intensity at the high energy side, which could be caused by an unresolved standing spin wave mode. There might be an additional broad peak around 400 meV (visible in Figs. 1 and 2(b)), especially at larger ΔK. In any case, if these standing spin wave branches are there, they have a much lower cross section compared to the acoustic mode.

The results above are discussed within the simple nearest neighbor Heisenberg model. For a quantitative description of the SPEEL spectra a theory is necessary, which takes into account the itinerant character of the electron spin in Co [22,23]. For Co such a theoretical calculation has been performed only for bulk HCP Co, where it agrees well with neutron data, but none for ultrathin FCC Co layers up to now [24,25]. For bulk FCC Co *first principles* calculations are available, which, however, use the adiabatic approach. Therefore, the influence of the Stoner excitations is neglected in these calculations [26,27]. Nevertheless, the calculated spin wave energies of Refs. [26,27] are in fair and good agreement, respectively, with our experimental data when scaled down to the thin film. (dived by $\frac{12}{8}$ in the nearest-neighbor Heisenberg model)

The width and the intensity of the spin-wave peak has been extracted from the gaussian fit as well and is plotted in Fig. 3(b). The measured width is corrected for the resolution of the instrument. Since the loss peaks deviate somewhat from a gaussian curve by a tail towards the high loss side, the spin-wave peak width may be slightly overestimated. Despite this uncertainty it is evident, that the spin-wave peak is significantly broadened. The intensity of the spin-wave peak strongly decreases towards higher wave vectors of the spin wave. (Note, that the spectra in Fig. 3 are normalized to the diffuse elastic peak intensity, which drops by more than 2 orders of magnitude with ΔK in the investigated range.)

The spin-wave excitation cross-section depends strongly on the energy E_0 of the incident electrons. In Fig. 4(a) a series of SPEEL spectra taken with different E_0 but with fixed $\Delta K = 0.7\ \text{Å}^{-1}$ are plotted. The spin-wave intensity has a clear maximum at low E_0 around $E_0 = 7$ eV. The spin wave intensity at higher E_0 is much smaller and no clear peak is observable in the intensity spectra in Fig. 4(a). However, the asymmetry spectrum at $E_0 = 25$ eV seems to indicate an increased spin-wave intensity with respect to $E_0 = 15$ eV.

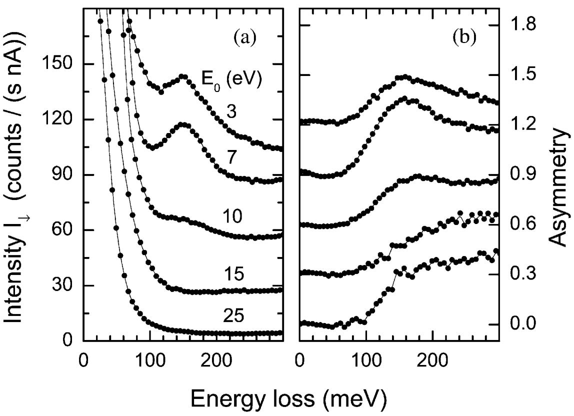

Fig. 4. SPEEL intensity spectra from 8 ML Co on Cu(0 0 1) for various energies E_0 of the incident electrons but at fixed $\Delta K = 0.7\ \text{Å}^{-1}$. $I_\downarrow$ intensity spectra, (b) asymmetry spectra calculated from $I_\downarrow$ and $I_\uparrow$ spectra.

3.2. 5 ML Fe on W(1 1 0)

Fig. 5 shows a series of SPEEL spectra from 5 ML Fe on W(1 1 0) along the $\langle 0 0 1 \rangle$-direction for $E_0 =$ 30.4 meV. The asymmetry in this case is much lower than for Co. Due to long measurement times the Fe surface is partially covered with adsorbates, which show up as vibration losses in the intensity spectra and small dips in the asymmetry spectra at about 130 and 250 meV. While for Co the spin-wave intensity was of the order of several percent of the intensity of the diffuse elastic peak, no such peaks are observable in the $I_\downarrow$ spectrum. Nevertheless, in the asymmetry spectrum a broad structure extending up to 500 meV is visible for all ΔK, which we attribute to spin-wave excitation. This structure does not show significant dispersion. Measurements at low E_0, as well as measurements on 3 ML Fe on 1 ML Co/Cu(0 0 1) gave qualitatively the same result.

One might attribute the different behavior of Fe and Co regarding the spin wave cross section to the fact, that Fe is a weak ferromagnet, i.e., the majority d-band is not completely filled, while Co is a strong ferromagnet, the majority d-band is completely filled and an energy gap of about 270–300 meV for d–d Stoner excitation within the d-band exists [6,28]. It is also known from neutron scattering that for Fe the spin wave broadens significantly for spin wave energies above ≈100 meV due to strong interaction with Stoner excitations [29,30]. For Co being a strong ferromagnet one would expect a much weaker interaction between spin waves and Stoner excitations. However, Penn and Apell [31] showed, that—at least for large energy losses—free-electron-like Stoner excitations are even more probable than d–d-

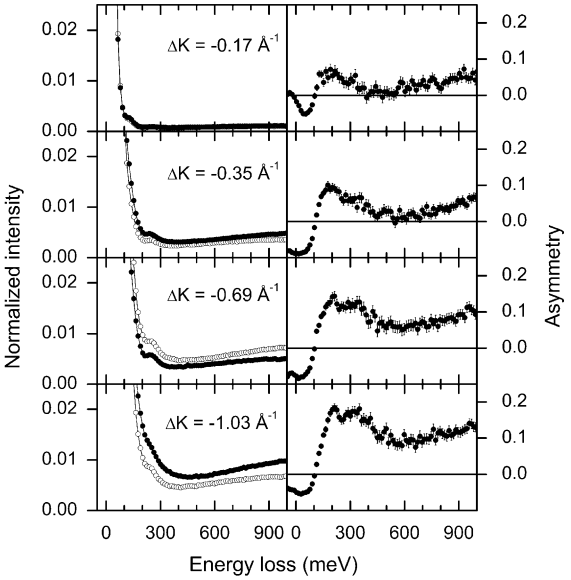

Fig. 5. Series of SPEEL intensity spectra from 5 ML Fe on W(1 1 0) along the ⟨0 0 1⟩ direction. Left panels: Intensities ● (○) for $I_{\uparrow(\downarrow)}$ normalized to the average intensity at 0 meV energy loss.

electron Stoner excitations. In fact, the calculations of Ref. [23] for 1 ML Fe on W(1 1 0) show sharp spin wave peaks only below ≈100 meV, while at higher energies they become very broad. For 5 ML Fe on W(1 1 0) the theoretical spectra at large ΔK are quite similar to what we observed [32]. SPEELS measurements with higher resolution are in preparation to explore the low energy range of spin wave excitation in Fe on W(1 1 0).

4. Conclusion

Spin-wave dispersion curves can be measured by Spin-polarized electron energy loss spectroscopy up to the surface Brillouin zone boundary. On FCC Co films on Cu(0 0 1) the spin-wave dispersion curve exhibits a clear thin film character and deviates significantly from the bulk like dispersion curve. The spin-wave cross-section in the SPEEL spectrum depends strongly on the incident energy. Contrary to Co, Fe films do not show a well defined spin wave peak in the SPEEL spectrum but a rather broad and weak feature extending up to 500 meV.

References

[1] B. Hillebrands, K. Ounadjela (Eds.), Spin dynamics in confined magnetic structures I, Topics in Applied Physics, Vol. 83, Spinger, Berlin, Heidelberg, New York, 2002.
[2] A. Schreyer, et al., J. Appl. Phys. 87 (2000) 5443.
[3] H. Hopster, R. Raue, R. Clauberg, Phys. Rev. Lett. 53 (1984) 695.
[4] J. Kirschner, D. Rebenstorff, H. Ibach, Phys. Rev. Lett. 53 (1984) 698.
[5] D. Venus, J. Kirschner, Phys. Rev. B 37 (1988) 2199.
[6] K.-P. Kämper, D.L. Abraham, H. Hopster, Phys. Rev. B 45 (1992) 14335.
[7] M. Plihal, D.L. Mills, J. Kirschner, Phys. Rev. Lett. 82 (1999) 2579.
[8] G. Vignale, K.S. Singwi, Phys. Rev. B 32 (1985) 2824.
[9] M.P. Gokhale, A. Ormeci, D.L. Mills, Phys. Rev. B 46 (1992) 8978.
[10] M. Plihal, D.L. Mills, Phys. Rev. B 58 (1998) 14407.
[11] H. Ibach, D. Bruchmann, R. Vollmer, M. Etzkorn, P.S. Anil Kumar, J. Kirschner, Rev. Sci. Instrum. 74 (2003) 4089.
[12] D.T. Pierce, et al., Rev. Sci. Instrum. 51 (1980) 778.
[13] P. Drescher, et al., Appl. Phys. A 63 (1996) 203.
[14] A. Clarke, et al., Surf. Sci. 187 (1987) 327.
[15] A.K. Schmid, et al., Phys. Rev. B 48 (1993) 2855.
[16] U. Gradmann, G. Waller, Surf. Sci. 116 (1982) 539.
[17] D. Sander, et al., J. Magn. Magn. Mater. 177 (1998) 1299.
[18] R.N. Sinclair, B.N. Brockhouse, Phys. Rev. 120 (1960) 1638.
[19] D.P. Pappas, et al., Phys. Rev. Lett. 66 (1991) 504.
[20] H. Hopster, J. Electron. Spec. Relat. Phenom. 99–98 (1999) 17.
[21] M. Grimsditch, A. Malozemoff, A. Brunsch, Phys. Rev. Lett. 43 (1979) 711.
[22] H. Tang, M. Plihal, D.L. Mills, J. Magn. Magn. Mater. 187 (1998) 23.
[23] R.B. Muniz, D.L. Mills, Phys. Rev. B 66 (2002) 174417.
[24] K.N. Trohidou, J.A. Blackman, J.F. Cooke, Phys. Rev. Lett. 67 (1991) 2561.
[25] T.G. Perring, A.D. Taylor, G.L. Squires, Physica B 213–214 (1995) 348.
[26] M. Pajda, et al., Phys. Rev. B 64 (2001) 174402.
[27] S.V. Halilov, H. Eschrig, A.Y. Perlov, P.M. Oppeneer, Phys. Rev. B 58 (1998) 293.
[28] C.M. Schneider, et al., J. Phys.: Condens. Matter 3 (1991) 4349.
[29] D. McKenzie Paul, P.W. Mitchell, H.A. Mook, U. Steigenberger, Phys. Rev. B 38 (1988) 580.
[30] T.G. Perring, et al., J. Appl. Phys. 69 (1991) 6219.
[31] D.R. Penn, P. Apell, Phys. Rev. B 38 (1988) 5051.
[32] D.L. Mills, Private communication.