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Propagation of Nd magnetic phases in Nd/Sm(001) superlattices

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Abstract

The propagation of Nd long range magnetic order in the hexagonal and cubic sublattices has been investigated in double hexagonal compact Nd/Sm(001) superlattices by resonant x-ray magnetic scattering at the Nd L_2 absorption edge. For a superlattice with 3.7 nm thick Sm layers, the magnetic structure of the hexagonal sublattice propagates coherently through several bilayers, whereas the order in the cubic sublattice remains confined to single Nd blocks. For a superlattice with 1.4 nm thick Sm layers, the magnetic structures of both sublattices appear to propagate coherently through the superlattice. This is the first observation (i) of the long range coherent propagation of Nd order on the cubic sites between Nd blocks and (ii) of a different thickness dependence of the propagation of the Nd magnetic phases associated with the hexagonal and cubic sublattices. The propagation of the Nd magnetic order through Sm is interpreted in terms of generalized susceptibility of the Nd conduction electrons.

1. Introduction

In rare earth metals, the different magnetic structures result from the competition between the long range indirect exchange interaction between the localized 4f moments through the conduction electrons, and the local crystal field. The magnetic behaviour is then very sensitive to slight modifications of the inter-atomic distances, e.g due to strains. Advances in molecular beam epitaxy (MBE) have allowed the production of artificial structures like superlattices, periodic stacks of single crystal bilayers, where these strains may be finely monitored by adjusting the components of the layers and their thicknesses. Moreover, superlattices present new physical properties correlated with the presence of a nanoscale period. One of the most spectacular phenomena in rare earth superlattices is the coherent propagation of the long range magnetic order of one of the superlattice components (A) through the other one (B), which may be magnetically ordered or not [1–6]: one could establish [7] that the long range magnetic order propagates from an individual layer A to the next one, via a spin density wave (SDW) in the conduction band of the B spacer layer. The limited extension and damping of the SDW in B accounts for a maximum critical thickness of the layers B above which the coherent propagation cannot establish. This is the so-called spacer critical thickness.

Coherent magnetic propagation depends on a number of conditions. In particular, both constituents of the superlattice should exhibit similar crystal and electronic structures, to enable the matching of Fermi surfaces at the interfaces. However, to determine whether coherent propagation might occur, a still debated issue is the relative importance of (i) the non-local magnetic response of the spacer layer B (its static susceptibility $\chi(q)$) and (ii) the matching of the wavefunctions and the band structures at the interfaces [8].

Long range magnetic order was first evidenced in hcp rare earth superlattices [1–5]. In systems with double hexagonal compact structure (dhcp), the presence of two sublattices of non-equivalent crystal sites, of hexagonal or cubic local symmetry, is an additional source of complexity. In dhcp rare earths (Nd, Sm, Ce), both sublattices order in a different magnetic structure, with different propagation vectors and different ordering temperatures.

The magnetic order of Nd in superlattices has already been studied in the Nd/Pr [6], Nd/La [9] and Nd/Ce [10] systems. The incommensurate magnetic structure of the hexagonal sublattice is similar to the bulk one and it propagates coherently across several bilayers in the three systems. Coherence has been evidenced with Pr spacer layers as thick as 33 atomic planes (≈10 nm) in Nd/Pr. The same studies have shown that the magnetic structure of the cubic sublattice in Nd/Pr and Nd/La superlattices is also the same as the bulk one, but does not propagate coherently through ten atomic layers (≈3 nm) of Pr or La. In Nd/Ce superlattices [10] with 30 atomic plane thick Nd layers (≈9 nm), the case is significantly different since the Nd cubic sublattice exhibits a c-axis ferromagnetic order, similar to the magnetic structure observed in bulk Nd under pressure. This ferromagnetic phase propagates coherently through the Ce spacer layers that are supposed to exhibit a similar ferromagnetic order.

We present here a study of the magnetism of Nd in Nd/Sm(001) superlattices, where both Nd and Sm exhibit the same double hexagonal compact crystal structure [11]. The initial interest in such systems is that one expects unusual effects, as, in their bulk phase, both components of the superlattice order with different magnetic structures, incommensurate in Nd and commensurate in Sm, and different magnetic moment directions, along $\mathbf{c}^*$ in Sm [11] and close to the basal plane in Nd [12–15]. Also, as established in rare earth superlattices, the same dhcp stacking for both constituents is a prerequisite to coherent propagation of magnetic orders. This system is thus well suited to study the propagation of Nd magnetic orders through magnetic spacer layers.

This paper concentrates on the magnetic order in the Nd layers. Section 2 summarizes the experimental details, while section 3 gives an overview of the results: several magnetic phases are evidenced in the cubic or the hexagonal sublattices and coherent propagation occurs with different critical thicknesses of the Sm spacer layers. Section 4 presents a general discussion of the conditions to be fulfilled to get coherent propagation.

2. Experimental details

Although bulk Sm exhibits a nine hexagonal plane stacking sequence, previous investigation devoted to pure Sm has shown that several hundred nanometre thick Sm films with a dhcp stacking sequence could also be obtained by MBE [11]. Taking advantage of this method, we could grow Nd/Sm(001) superlattices, deposited onto a (110) sapphire substrate covered with a 80 nm thick Nb(110) buffer. This paper reports on the results obtained in two Nd/Sm(001) superlattices: $[\mathrm{Nd}(18.8\ \mathrm{nm})/\mathrm{Sm}(3.7\ \mathrm{nm})]_{42}$ and $[\mathrm{Nd}(15.4\ \mathrm{nm})/\mathrm{Sm}(1.4\ \mathrm{nm})]_{43}$, referred to as S1 and S2, respectively. The subscripts correspond to the number of bilayer repeats. From x-ray diffraction, both elements Nd and Sm exhibit the same dhcp stacking sequence along the growth direction and the dhcp stacking is coherent over many bilayers [11].

The long range magnetic order in the Nd layers has been studied using resonant x-ray magnetic scattering (RXMS). This technique is chemically selective, which makes it particularly well suited to the study of such binary systems as superlattices, and offers high resolution in the reciprocal space. It allows one to detect the coherence of a magnetic structure through a superlattice, and measure coherence lengths on the scale of atomic distances. More precisely, the signature of the coherence of a magnetic structure through several bilayers is the splitting of the magnetic Bragg peaks into an array of peaks separated by a distance c/Λ along the growth direction (**c** in our case), where Λ is the period of the superlattice (bilayer thickness). The magnetic coherence length ξ along the **c** direction is defined as the size of blocks where long range magnetic order is established, and can be deduced from the (resolution corrected) full width at half maximum of the magnetic peaks in the growth direction ΔQ: $\xi \approx 2\pi/\Delta Q$ (with ΔQ in Å^{-1}).

The RXMS experiments were carried out at the ID20 beamline (European Synchrotron Radiation Facility, in Grenoble, France), with polarization analysis of the scattered beam, using the (220) reflection from a Cu crystal, to reduce the charge background.

The incident photon beam energy was tuned to the Nd L_2 absorption edge at E = 6.723 keV where the resonant intensities are about one order of magnitude stronger than at the L_3 edge [16], due to both a branching ratio of the order of six, and a lower absorption by the windows and air gaps along the beam path. In the Nd/Sm(001) superlattices, the magnetic phases in Sm layers have been shown to be very close to the ones evidenced in dhcp thick films [17].

3. Results

We first present the magnetic structures observed in the Nd layers, before we consider their propagation through the Sm spacer layers. In the following, the scattering vector is defined as $\mathbf{Q} = (Q_H\, Q_K\, Q_L)$ in the $(\mathbf{a}^*\mathbf{b}^*\mathbf{c}^*)$ basis.

3.1. Magnetic structure in the Nd layers

For both samples, several magnetic reflections corresponding to the incommensurate order usually associated with the hexagonal sublattice in bulk Nd have been evidenced below 20 K. They appear around (004) and (008) charge reflections, at positions $(\tau_{\text{Hhex}}\ 0\ Q_L)$ with Q_L = 4 ± 1 and 8 ± 1, in agreement with the bulk magnetic propagation vector $\boldsymbol{\tau}_{\text{hex}} = (\tau_{\text{Hhex}}\ 0\ 1)$ with $0.126 < \tau_{\text{Hhex}} < 0.14$ [13]. At 8 K, additional magnetic peaks appear at positions $(\tau_{\text{Hcub}}\ 0\ Q_L)$ and are attributed to magnetic order in the cubic sublattice by analogy with bulk Nd [13] where the propagation vector is $\boldsymbol{\tau}_{\text{cub}} = (\tau_{\text{Hcub}}\ \varepsilon\ 1)$, with ε vanishing below 7 K.

As an illustration, figures 1 and 2 show the thermal variation of the in-plane components $\tau_{\text{H}}(=\tau_{\text{Hhex}}$ or $\tau_{\text{Hcub}})$ of the magnetic propagation vectors, and of the integrated intensity, measured at the $(\tau_{\text{H}}\ 0\ 7)$ magnetic peaks. The solid lines in figure 1 correspond to bulk Nd behaviour [13]. The Néel temperatures and propagation vectors in both samples are close to the bulk ones. However, on cooling, the τ_{Hhex} values shift slightly away from the bulk values and the τ_{H} splitting observed in bulk Nd at 6 K is missing. The biggest discrepancies with the bulk behaviour occur at low temperature: the intensities of the peaks attributed to the magnetic order in the hexagonal and cubic sublattices decrease rapidly below 5 K for S1 or 4 K for S2, and vanish around 3 K for S1 and 2 K for S2. Simultaneously, new peaks, corresponding to a new magnetic phase, appear at intermediate Q_{H} values: Q_{Hnew} = 0.1525 and 0.158 rlu (S1)

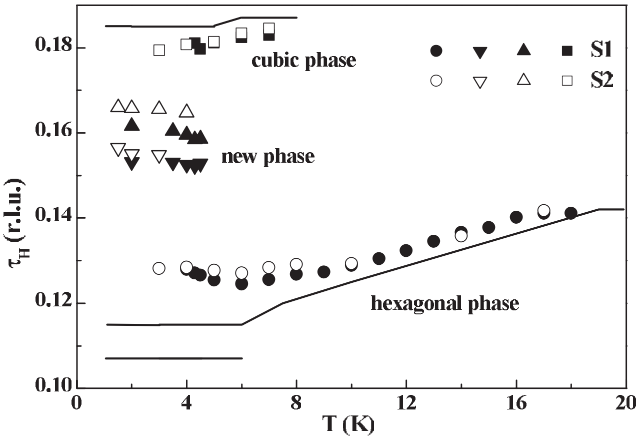

Figure 1. Thermal variation of the τ_H component of the magnetic propagation vector for the Nd hexagonal (circles) and cubic sublattice (squares) and for the new low temperature phase (up and down triangles) for S1 (full symbols) and for S2 (open symbols). Bold lines are bulk values [13].

and $Q_{\mathrm{Hnew}} = 0.1525$ and 0.164 rlu (S2). This unusual Nd phase could be associated with a common magnetic order of the hexagonal and cubic sites [17]. Such a structure has never been observed in other Nd based superlattices [6, 7, 9, 10].

3.2. Propagation of the Nd magnetic order through the Sm spacer layers

To study the propagation of the Nd magnetic phases across the Sm spacer, we performed scans of the scattering vector along **c*** across the magnetic reflections associated with hexagonal, cubic and new magnetic orders. Typical scans are shown in figure 3, for $Q_L = 7$ rlu.

For the hexagonal sublattice, (figures 3(a) and (b)), we observe two magnetic peaks separated by c/Λ, characteristic of a long range coherent propagation. Their width is similar to the width of the structural peaks. The deduced coherence length, $\xi \approx 70$ nm, shows that the magnetic structure propagates through several bilayer repeats in both samples. The critical thickness of the Sm layer, below which coherent propagation of Nd structure is allowed is thus larger than 3.7 nm for the Nd hexagonal sublattice. Moreover, the presence of two magnetic peaks is a signature of the occurrence of different magnetic orders in Nd and Sm layers (i.e. of a magnetic contrast between the Nd and Sm layers).

For the cubic sublattice, the line shape for S2 (figure 3(d)) also shows two narrow peaks corresponding to a coherence length $\xi \approx 67$ nm. For S1, however, a single broader peak is observed (figure 3(c)), corresponding to a coherence length $\xi \approx 17$ nm, slightly smaller than the thickness of a single Nd layer. For the propagation of the Nd cubic magnetic order, the critical thickness of the Sm layer is thus comprised between 1.4 and 3.7 nm.

Finally, for the new phase (figures 3(e) and (f)), coherent propagation through several bilayers is also observed. At low temperature $\xi \approx 60$ nm in S1. It reaches $\approx$80 nm in S2, where the Sm spacer layers are thinner.

The temperature dependences of the coherence lengths in the various Nd magnetic phases are summarized in figure 4 for both samples. The magnetic coherence length of the order in the Nd hexagonal sublattice, which is the first to establish on cooling, is constant above 5.5 K (S1) and 7 K (S2). It is similar in both samples, despite different Sm spacer thicknesses. The onset of the order in the cubic sublattice does not seem to affect the coherence length of the order in the hexagonal sublattice. In S1, the coherence length of the cubic magnetic order remains small. In contrast, in S2, it increases from 25 nm at the ordering temperature to 67 nm at 5 K. At the transition to the new phase, while the higher temperature phases vanish, the coherence length of those structures which propagate through the superlattices (hexagonal phase for S1

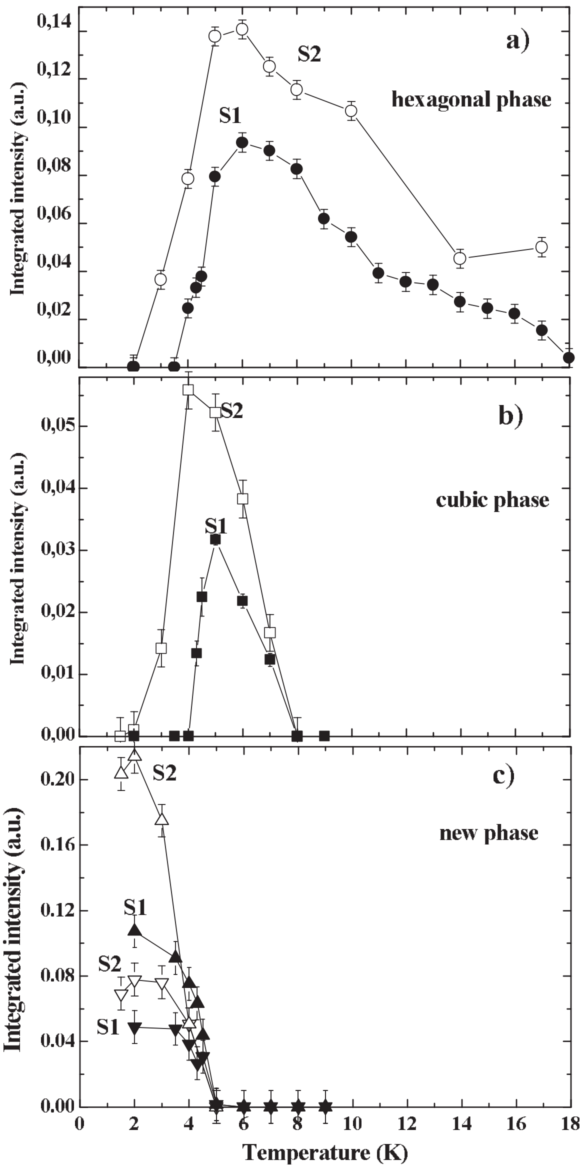

Figure 2. Thermal variation of integrated intensities of the magnetic peaks attributed to the ordering on the Nd hexagonal sites (circles, (a)), on the Nd cubic sites (squares, (b)) and to the new low temperature phase (up and down triangles, (c)) for S1 (full symbols) and for S2 (open symbols). The lines are guides for the eyes.

and cubic and hexagonal ones for S2) decreases. For the cubic phase, this loss of coherence may be slightly delayed.

4. Discussion

The main result of this paper concerns the different propagations of the magnetic phases in the hexagonal and cubic sublattices throughout Nd/Sm superlattices. In S1, with 3.7 nm thick Sm layers, only the magnetic order of the hexagonal sublattice propagates coherently through the Sm layers, while the order in the cubic sublattice remains confined in the Nd individual layers. In S2, with thinner Sm layers (1.4 nm), the magnetic structures of both the hexagonal and the cubic sublattices propagate simultaneously and coherently through Sm.

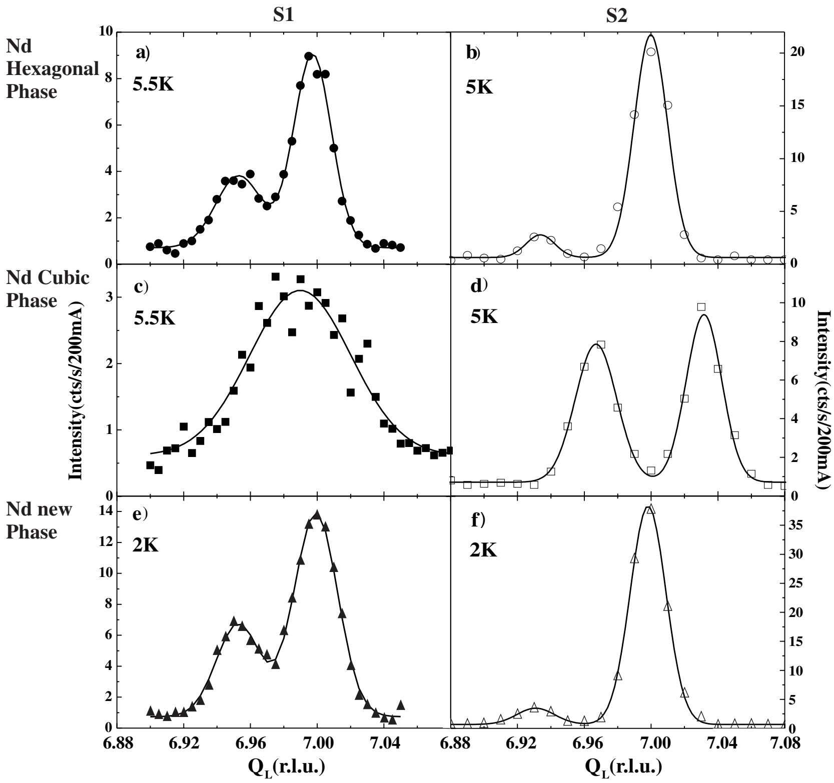

Figure 3. Scans of **Q** in the **c*** direction measured across the various magnetic peaks for S1 (left column) and for S2 (right column). (a) (0.125 0 Q_L) at 5.5 K; (b) (0.127 0 Q_L) at 5 K; (c) (0.18 0 Q_L) at 5.5 K; (d) (0.18 0 Q_L) at 5 K; (e) (0.162 0 Q_L) at 2 K; (f) (0.166 0 Q_L) at 2 K. Intensities are normalized to the maximum ring current of 200 mA.

This is the first observation of coherent propagation of the modulated structure of the Nd cubic sublattice, showing that magnetic orders in both sublattices can propagate coherently and simultaneously. The Sm critical thicknesses are significantly different for the magnetic structures in the hexagonal and cubic Nd sublattices.

As already mentioned in the introduction, studies of dhcp Nd/Pr [6] have shown that the long range magnetic order of the Nd hexagonal sublattice could propagate coherently through Pr layers as thick as 33 atomic planes. Since the extension of exchange between Nd hexagonal sites over 33 atomic planes (10 nm) seems much larger than expected from Pr generalized susceptibility, the authors argue that the Fermi surface-matching mechanism should account for the coherence observed in the hexagonal sublattice. This argument does not account for the absence of propagation of the cubic magnetic structure, either through Pr [6] or through La [9], even with a 3 nm (10 atomic layers) thin spacer.

In Nd/Sm superlattices, the coherent propagation of the cubic magnetic structure is achieved for thin enough spacer layers (approximately five atomic planes). This phenomenon may be surprising because of the magnetic character of the Sm layers with competing anisotropy compared to Nd, but their small thickness makes the propagation possible.

The existence of different spacer critical thicknesses for both sublattices is a key point in this study. It points towards intrinsic differences between these two magnetic orders in Nd and

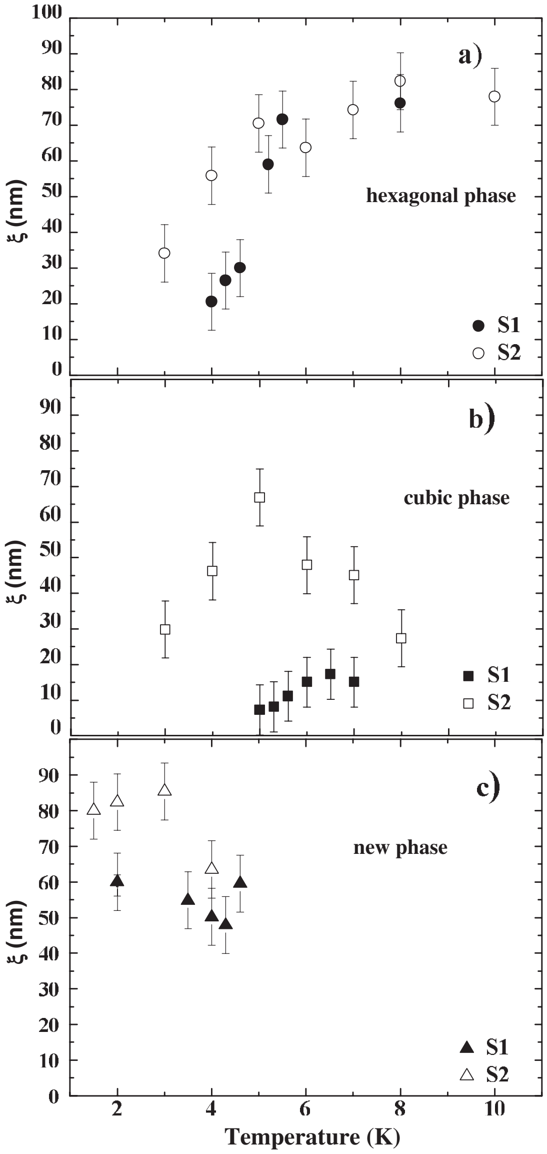

Figure 4. Thermal variation of the magnetic coherence length for hexagonal (a) and cubic (b) sublattices and for the new magnetic phase (c) for S1 (filled symbols) and for S2 (open symbols).

suggests that, beyond the role of conduction electron susceptibility in the Sm spacer layer, the Nd intrinsic electronic properties must be considered to account for the propagation.

Experimental data on $\chi(\mathbf{Q})$ are not available for Nd metal. However, bulk Nd and Pr have similar magnetic structures at the ordering temperature, and one can assume, as a first approximation, that they exhibit similar Fermi surfaces and magnetic excitation spectra. The measured excitation spectra of Pr metal provide evidence for a susceptibility of the conduction electrons that is large for an antiferromagnetic configuration of the hexagonal layers and small for cubic site ordering [18]. From Houmann *et al* [19], the dispersion relation near zero wavevector varies rapidly on the Pr hexagonal sites, indicating a very long range interaction. In contrast, the excitations propagating in the Pr cubic sublattice have negligible dispersion, indicating that the total coupling between different atomic planes of cubic ions is very small.

Expected similar features in Nd justify that Nd generalized susceptibility is different for an antiferromagnetic configuration between hexagonal or between cubic different atomic planes, and consequently, that the Nd cubic ordering cannot propagate through as thick a spacer layer as the hexagonal ordering does.

This intrinsic limitation to the coherent propagation of the magnetic order in the cubic sublattice may also explain the previous results obtained in other Nd based superlattices. The Pr and La spacer thicknesses used in these studies are most probably lying in between the two critical thicknesses: below the threshold for the Nd hexagonal sublattice but above for the cubic one.

5. Conclusion

This study of dhcp Nd/Sm superlattices has brought the first evidence of coherent propagation of the Nd magnetic structure of the cubic sublattice, through 1.4 nm thick Sm layers. This coherent phase coexists with the coherent order of the hexagonal sublattice, while only the order in the hexagonal sublattice propagates coherently through 3.7 nm thick Sm layers. The existence of two different critical thicknesses of the Sm spacer enlightens the importance of the conduction electron susceptibility of Nd in the mechanism responsible for the propagation of magnetic order in these Nd based superlattices. These results thus provide important information for a better understanding of magnetic interactions in magnetic multilayers.

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References

[1] Erwin R W, Rhyne J J, Salamon M B, Borchers J, Sinha S, Du R R, Cunningham J E and Flynn C P 1987 *Phys. Rev.* B **35** 6808
[2] Borchers J A, Salomon M B, Erwin R W, Rhyne J J, Du R R and Flynn C P 1991 *Phys. Rev.* B **43** 3123
[3] Jehan D A, McMorrow D F, Cowley R A, Wells M R, Ward R C C, Hagman N and Clausen K N 1993 *Phys. Rev.* B **48** 5594
[4] Dumesnil K, Dufour C, Mangin Ph, Marchal G and Hennion M 1996 *Phys. Rev.* B **54** 6407
[5] Simpson J A, Cowley R A, Jehan D A, Ward R C C, Wells M R, McMorrow D F, Clausen K N, Thurston T R and Gibbs D 1996 *Z. Phys.* B **101** 35
[6] Goff J P, Bryn-Jacobsen C, McMorrow D F, Ward R C C and Wells M R 1997 *Phys. Rev.* B **55** 12537
[7] Goff J P, Sarthour R S, McMorrow D F, Yakhou F, Stunault A, Vigliante A, Ward R C C and Wells M R 1999 *J. Phys.: Condens. Matter* **11** L139
[8] Cowley R A 1998 *J. Magn. Magn. Mater.* **177** 1156
[9] Goff J P, Sarthour R S, McMorrow D F, Rainford B D, Wilkins C J T, Ward R C C and Wells L R 1997 *Physica* B **241–243** 714
[10] Clegg P S, Goff J P, McIntyre G J, Ward R C C and Wells L R 2003 *Phys. Rev.* B **67** 174414
[11] Dufour C, Dumesnil K, Soriano S, Pierre D, Senet Ch and Mangin Ph 2002 *J. Cryst. Growth* **234** 447
[12] Moon R M, Koehler W C, Sinha S K, Stassis C and Kline G R 1979 *Phys. Rev. Lett.* **43** 62
[13] Lebech B, Wolny J and Moon R M 1994 *J. Phys.: Condens. Matter* **6** 5201
[14] Forgan E M, Gibbons E P, McEwen K A and Fort D 1989 *Phys. Rev. Lett.* **62** 470
[15] Dufour C, Dumesnil K, Soriano S, Mangin Ph, Brown P J, Stunault A and Bernhoeft N 2002 *Phys. Rev.* B **66** 94428
[16] Watson D *et al* 1996 *Phys. Rev.* B **53** 726
[17] Soriano S, Dufour C, Dumesnil K and Stunault A 2006 *J. Phys.: Condens. Matter* at press
[18] Jensen J and MacIntosh A R 1991 *Rare Earth Magnetism* (New York: Oxford University Press)
[19] Houmann J G, Rainford B D, Jensen J and MacIntosh A R 1979 *Phys. Rev.* B **20** 1105