Refinement of the crystal field interaction for the rare earth intermetallic series $RNiAl_4$ (R = rare earth)

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The intermetallic series $RNiAl_4$ (R = rare earth) exhibits interesting magnetic behaviour [1-2], including the potential for low temperature, inverse, magnetic cooling [3]. Given that the $RNiAl_4$ magnetism is associated solely with the R sub-lattice and is influenced strongly by the local crystal field (CF) interaction at the R-site, it is important that the CF interaction be characterised. Thermal neutrons were employed here in an effort to extend a previous cold neutron inelastic neutron scattering (INS) investigation [4] to higher energy CF transitions at the single Er^{3+} site in $ErNiAl_4$.

Substantial amounts of $ErNiAl_4$ (34.8 g) and $YNiAl_4$ (26 g) were prepared as a set of smaller 1-2 g lots via repeated argon arc melting followed by vacuum annealing for 5-6 d at 1050 °C. Neutron scattering measurements were performed using both the elastic and inelastic modes of operation of the C5 polarised triple-axis spectrometer at CNBC. The INS spectra were accumulated with a final scattering energy of E_f = 14 meV.

The elastic neutron diffraction pattern (λ = 2.37051 Å) recorded for $ErNiAl_4$ at 3.9 K was consistent with an incommensurate sinusoidal magnetic structure, a propagation vector of [0.191 1.0 0.038] and a local moment amplitude of $\mu(Er^{3+}) = 7.0\ \mu_B$ aligned with the c-axis. However, recent ^{166}Er-Mossbauer results[5] rule out a spread in the local Er moment so that a square wave modulation is likely more appropriate. Using polarised neutrons (p // Q) with both spin flip (SF) and non spin flip (NSF) detection, the temperature dependence of the net magnetic intensity for the prominent reflection at $2\theta \approx 51.75°$ indicated an ordering temperature of 6.5 K (Fig. 1). This is in close agreement with the bulk specific heat value of T_N = 5.8 K [7].

Unpolarised INS spectra were recorded for $ErNiAl_4$ at 10 K with four different scattering vectors, Q, over the energy ranges of 0 - 25 meV (Q = 1.2 $Å^{-1}$), 0 - 41 meV (Q = 2.7 $Å^{-1}$), 0 - 50 meV (Q = 3.0 $Å^{-1}$), and 20 - 50 meV (Q = 5.2 $Å^{-1}$). As shown in Fig. 2, strong transitions were observed at 3.08(2), 7.53(2) and 11.6(1) meV. These lower energy transitions are in excellent agreement with those observed previously using cold neutrons [4] and the evident Q-independence confirms that they are associated with "magnetic" CF transitions.

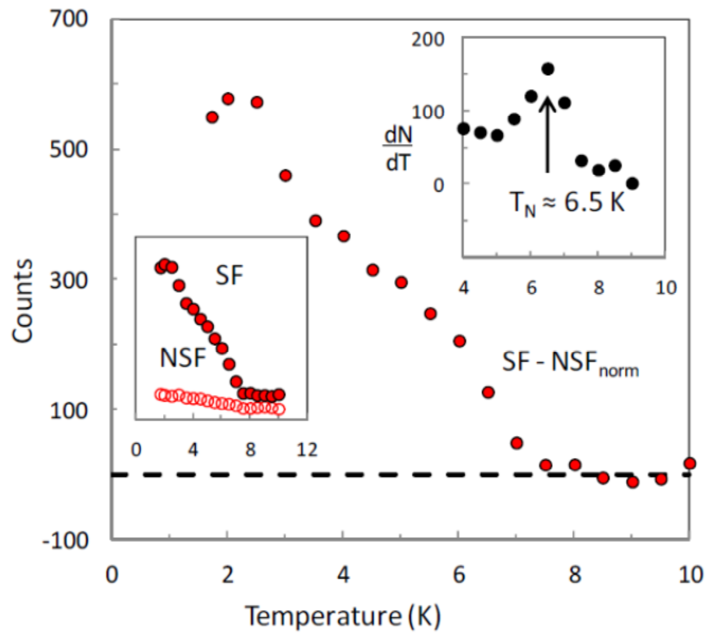

Figure 1 Temperature dependent intensity of the $ErNiAl_4$ magnetic reflection at $2\theta = 51.75°$ (λ = 2.37051 Å): SF = spin flip, NSF = non spin flip, and NSF_{norm} indicates NSF normalised to SF in the temperature range of 7.5 < T < 10 K.

From Fig. 3, the INS spectra for the higher energy range of 15 - 50 meV are seen to be of considerably lower intensity with broad features that are assumed to be associated with phonon transitions. In a first approach to separate out the true CF transitions, additional INS spectra were recorded for non-magnetic $YNiAl_4$ where there is no CF interaction. The intensities of the $YNiAl_4$ spectra were then scaled up so that their broad features matched as closely as possible those of their $ErNiAl_4$ counterparts and subtracted from the $ErNiAl_4$ spectra. Possible additional CF transitions were then identified at 15.4 and 18.2 meV as indicated by the superimposed triangular peaks in Fig. 3. A further candidate is observed at 34.2 meV with Q = 2.7 $Å^{-1}$ but it is no longer evident for Q = 3.0 and 5.2 $Å^{-1}$.

In a second approach, additional INS spectra were recorded for the higher energy range using polarised incident neutrons (for both SF and NSF). These data remain to be processed. Hopefully their statistical quality will prove sufficient for the broad phonon features to be stripped away via this alternative approach.

A more detailed description of the data analysis performed so far has been submitted for publication in the Proceedings of the 38th Annual Condensed Matter & Materials Meeting, Auckland (Waiheke Island), New Zealand, 4 - 7 Feb, 2014.

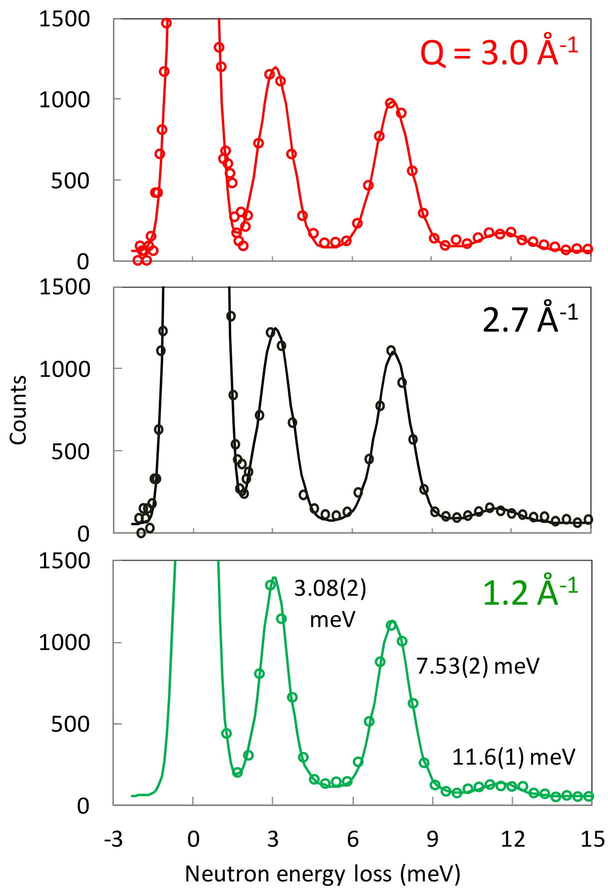

Figure 2 The 0 < E < 15 meV region of the INS spectra recorded for ErNiAl4 at 10 K with scattering vectors of Q = 1.2, 2.7 and 3.0 Å-1. The solid lines are fitted Pseudo-Lorentzian peaks superimposed on a linear background.

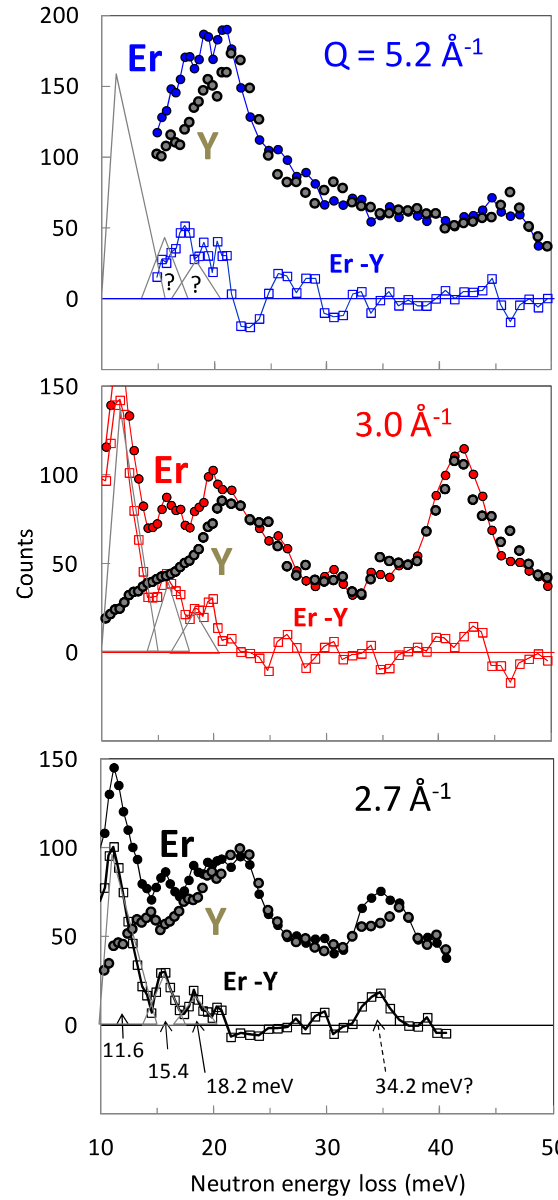

Figure 3 The 10 < E < 50 meV region of INS spectra recorded at 10 K with scattering vectors of Q = 2.7, 3.0 and 5.2 $Å^{-1}$. The data for $ErNiAl_4$, the scaled up data for $YNiAl_4$, and their subtraction are indicated by the labels "Er", "Y" and "Er - Y", respectively.

References

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