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LETTER TO THE EDITOR

Evidence for a magnon energy gap in the spin-slip phase of holmium

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Abstract. We have carefully re-measured the spin-wave dispersion relationship of holmium along $[00l]$ in its spin-slip $b = 11$ phase using a neutron triple-axis spectrometer. Our results are consistent with the existence of an energy gap in the spin-wave dispersion relationship close to the midpoint of the magnetic Brillouin zone. The measured magnitude of the gap of $\simeq 0.6$ meV is in good agreement with the value predicted for a spin-slip model of holmium by Jensen in 1988.

Although the basic magnetic structure of the heavy rare-earth metals has been known since the mid-sixties, recent diffraction experiments have revealed a wealth of unexpected detail which, in the case of holmium, has led to the development of the spin-slip model. The effect of spin slips on the magnetic excitations in holmium was shown by Jensen (1988) to be dramatic, producing gaps in the spin-wave dispersion at points in reciprocal space corresponding to the distance between spin slips. Here we report the first evidence for the existence of such a gap in the most fundamental spin-slip structure, the so-called one-spin-slip structure.

The pioneering neutron diffraction experiments by Koehler *et al* (1966) established that below $\simeq 133$ K holmium has a magnetic spiral structure in which the magnetic moments are bunched around the six easy directions in the basal plane. More recent diffraction studies on holmium using x-rays by Gibbs *et al* (1985) and neutrons by Cowley and Bates (1988) have shown that, while this model of the magnetic structure is correct in essence, it could not account for new features observed in the diffraction patterns. In the new model proposed by these authors below $\simeq 90$ K the magnetic spiral is composed of commensurate blocks of b unit cells of the low temperature cone structure separated by a regular array of discommensurations or spin slilps. As the temperature is decreased, b increases and the structure 'squares up' until it locks into a commensurate cone with a wavevector $\boldsymbol{q}_0 = \boldsymbol{c}^*/6$ at $\simeq 17.8$ K. Contained within the thermal evolution of the spin slips there are several structures which have on average a spin-slip spacing of $b = 3n - 1$, where n is an integer. In these special structures, the spin slips line up with each other and effectively alter the symmetry of the magnetic structure relative to that of the host lattice. This symmetry breaking has been shown to account for anomalous ultrasonic coupling effects observed in holmium (Bates *et al* 1988).

The effect on the spin waves of a simple pair-wise bunching of the magnetic moments about the easy directions, as originally proposed for the magnetic structure of holmium by Koehler *et al* (1966), was investigated in the alloy $Ho_{90}Tb_{10}$ by Larsen *et al* (1987). (This alloy was chosen for study by Larsen *et al* (1987) because the terbium ions confine the holmium moments to the basal plane, and also because it has a simple commensurable helical structure between 30 K to 20 K.) Their calculation predicted, amongst other interesting effects, the existence in the helical phase of an energy gap in the magnon dispersion relation at the mid-point of the magnetic Brillouin zone due to an effective doubling of the magnetic unit cell in the helical coordinate system. However, they were unable to resolve the two modes experimentally and concluded that the gap is probably less than 0.5 meV.

More recently Jensen (1988) has calculated the spin waves in holmium, assuming a spin-slip model for its magnetic structure. Although he presents results for both the conical phase and at several temperatures in the helical phase, we will only consider his results for the 'one-spin-slip' structure in the helical phase which has a number of interesting features, including the following: it has a net moment; is stable over a range of temperatures (Gibbs *et al* 1985); and it has an effective monoclinic symmetry which leads to anomalous ultrasound propagation (Bates *et al* 1988). This structure is so named because it has just one spin slip for each repeat distance of the ideal commensurate spiral. The number of lattice planes between spin slips is thus 11 and is referred to in the notation of Cowley and Bates (1988) as a $b = 11$ configuration. For the one-spin-slip structure, Jensen (1988) shows that, although energy gaps are expected to occur with a separation in reciprocal space of $\frac{1}{11}$, only the one corresponding to a wavevector of $q = \frac{5}{11}$ with respect to the magnetic zone centre is significant in his calculations, which have a resolution of 0.2 MeV. Moreover, the calculated value of the energy gap for $q = \frac{5}{11}$ is $\simeq$0.5 MeV, and is roughly twice the value calculated for the energy gap in the structure without spin slips (Larsen *et al* 1987). Previous measurements of the spin-wave dispersion relationship in pure holmium by Stringfellow *et al* (1970) did not report any such energy gap probably because their resolution of $\simeq$0.75 meV was too large. In this letter we report the results of a neutron inelastic scattering experiment with greatly improved resolution which confirms the existence of the energy gap at $q = \frac{5}{11}$.

We emphasise that our intention was not to re-measure the entire spin-wave branch along $[00l]$, but rather to focus our attention on that part of the branch where the gap was predicted to occur. Our experiments were mainly performed at Risø National Laboratory using the triple-axis spectrometer TAS-6 which is situated on the cold source at the DR3 reactor. (A preliminary study of the spin waves was made on the triple-axis neutron spectrometer IN12 at the Institut Laue–Langevin, Grenoble using the same small (0.1 cm^3) sample as had previously been characterised by Cowley and Bates (1988). This experiment, while encouraging, indicated that a much larger sample was required for the study to be completed in a reasonable time.) The single crystal of holmium used in our experiments was grown at the Ames laboratory and had a resistance ratio of $\simeq$130. It was in the form of a platelet $\simeq 30 \times 12 \times 4$ mm^3 with the c axis perpendicular to the largest face and was mounted in a variable temperature cryostat with the $(h0l)$ plane horizontal. Standard pyrolytic graphite monochromator and analyser crystals were used with horizontal collimations from reactor to detector of 60′–23′–22′–66′. The spectrometer was operated with the incident neutron energy held fixed at 8.3 meV.

The experiments were performed with wavevector transfers, $\boldsymbol{Q}$, between the 001 and 002 nuclear Bragg peaks and using the magnon branch originating from the 002-q

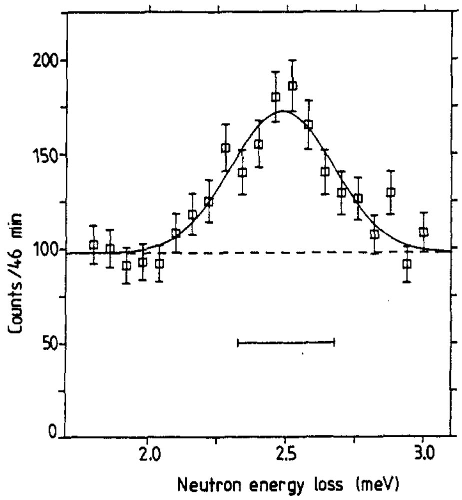

Figure 1. The measured scattered neutron energy distribution from holmium in its $b = 11$ phase ($T = 19.85$ K) obtained in a constant-$\boldsymbol{Q}$ scan with $\boldsymbol{Q} = (0, 0, 1.30)$ corresponding to $q = 0.52$. The full curve is the result of a least-squares fit to a Gaussian in addition to flat background (broken line). The fitted centre and width (FWHM) of the peak are respectively 2.48 (0.02) meV and 0.43 (0.05) meV. The horizontal bar is the measured vanadium FWHM for the $(1, -1, -1)$ configuration of the spectrometer used in this scan.

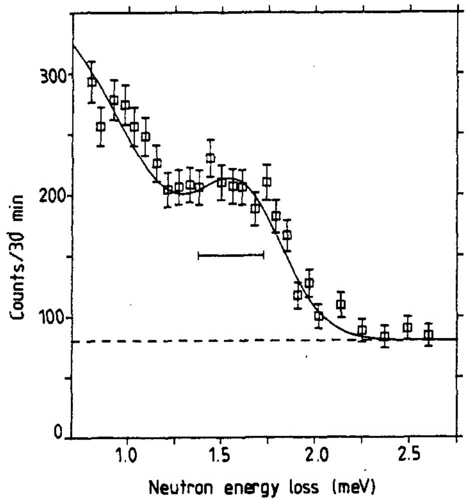

Figure 2. The measured scattered neutron energy distribution from holmium in its $b = 11$ phase ($T = 19.85$ K) obtained in a constant-$\boldsymbol{Q}$ scan with $\boldsymbol{Q} = (0, 0, 1.45)$, corresponding to $q = 0.37$. The full curve is the result of a least-squares fit to two Gaussians, one representing the quasi-elastic scattering and the other spin-wave scattering, in addition to a flat background (broken line). The fitted centre and width (FWHM) of the peak are respectively 1.61 (0.02) meV and 0.53 (0.03) meV. The horizontal bar is the measured vanadium FWHM for the $(1, -1, -1)$ configuration of the spectrometer used in this scan.

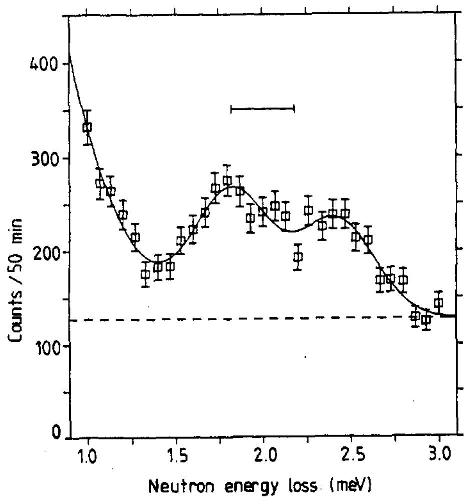

Figure 3. The measured scattered neutron energy distribution from holmium in its $b = 11$ phase ($T = 19.85$ K) obtained in a constant-$\boldsymbol{Q}$ scan with $\boldsymbol{Q} = (0, 0, 1.375)$, corresponding to $q = 0.44$. The full curve is the result of a least-squares fit to three Gaussians, one representing quasi-elastic scattering and the other two scattering by spin waves, in addition to a flat background (broken line). From the fit, the low energy peak is centred on 1.82 (0.03) meV with a width (FWHM) of 0.51 (0.05) meV, and the high energy peak is centred on 2.41 (0.03) meV with a width of 0.51 (0.03). The horizontal bar is the measured vanadium FWHM for the $(1, 1, 1)$ configuration of the spectrometer used in this scan.

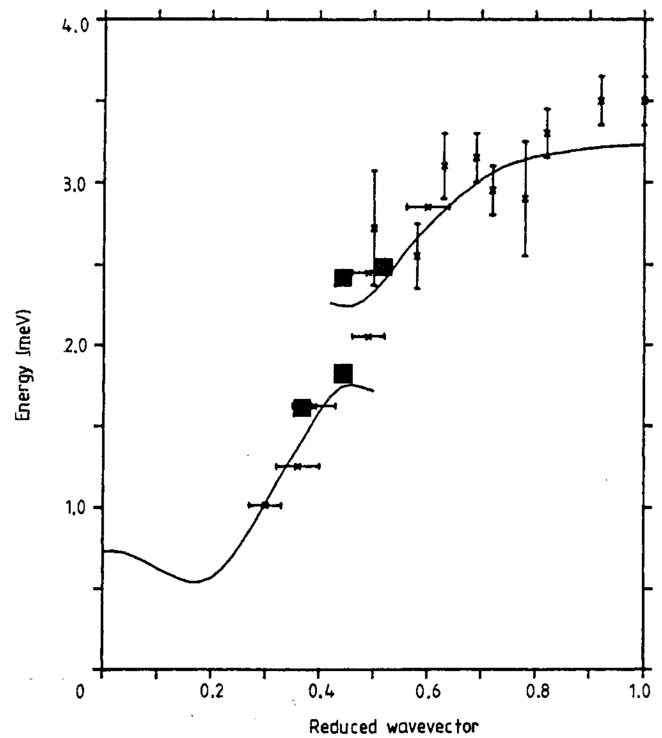

Figure 4. The spin-wave dispersion relation along $[0 0 l]$ of holmium. Full curve: from Jensen (1988); crosses: from Stringfellow *et al* (1970); full squares: from the fits to our experimental data shown in figures 1–3. We estimate that the errors in our data are comparable to the size of the symbols. The abscissa is in units of $2\pi/c$.

magnetic Bragg reflection. In order to minimise any instrumental broadening it is necessary to match as closely as possible the resolution function of the triple-axis spectrometer to the dispersion of the excitation under investigation. Away from the gap the average dispersion of the spin waves along $[0 0 l]$ for the particular zone in which we were working is -5 meV Å^{-1} and the most suitable configuration for the spectrometer was $(+1, -1, -1)$. At the wavevector transfer of the gap the dispersion of the spin waves is expected to be zero, and the vanadium width would then be largely independent of the spectrometer configuration. We decided to use the de-focused (1, 1, 1) setting of the spectrometer to degrade our Q-resolution, thereby increasing the count rate. (Here $+1$ (-1) indicates scattering to the left (right) of the straight through beam.) The measured vanadium widths (FWHM) were found to be, for the $(+1, +1, +1)$ and $(+1, -1, -1)$ configurations, respectively 0.36(3) meV and 0.35(3) meV.

The sample temperature was slowly decreased from above T_N and the value of b monitored by observing the positions of the first-order spin-slip peaks around the $10q$ magnetic Bragg reflection. The sample was stabilised at a temperature of 19.85 K which gave $b = 11.0(1)$ and allowed to equilibrate at this temperature for a few hours so as to avoid any problems with relaxation effects (Cowley and Bates 1988).

The scattered neutron energy distribution was measured by performing constant-Q scans at several positions on the dispersion curve. The results of three such scans made at reduced wavevectors of $q = 0.52$, 0.37 and 0.44 (i.e. either side and through the middle of the proposed gap) are shown respectively in figures 1–3.

In the scan through the spin-wave dispersion relation on the high energy side ($q = 0.52$) of the expected position of the gap a sharp, nearly resolution limited peak was observed (see figure 1). The centre of this peak, as determined by fitting the measured lineshape with a Gaussian, was in good agreement with the previous low resolution measurements of the spin waves in holmium by Stringfellow *et al* (1970). On the lower energy side ($q = 0.37$) of the gap we also observed a single peak in the scattered neutron energy distribution superimposed on a sharply sloping background (see figure 2). This sloping background was quasi-elastic in nature and found to be significant up to a neutron energy transfer of $\simeq 1.5$ meV. In contrast to the results given in figures 1 and 2, a scan with the wavevector transfer set at the expected position ($q = 0.44$) of the gap produced a scattered neutron distribution that was significantly broader than the measured resolution (see figure 3). Moreover, this distribution was consistent with two modes separated by $\simeq 0.6$ meV, each with a width close to the instrumental resolution. We believe that these two modes are the two branches of the spin waves on either side of the energy gap predicted by Jensen (1988).

In figure 4 we reproduce the dispersion relationship calculated by Jensen (1988) for the $b = 11$ structure. On this diagram we have plotted the positions of the peaks shown in figures 1 to 3 in addition to the data given by Stringfellow *et al* (1970). From this diagram it is clear that our measurements of the position and magnitude of the energy gap in the spin waves are in good agreement with the calculations by Jensen (1988). The data point of Stringfellow *et al* (1970) in the middle of the gap is presumably due to the poor resolution used in their experiments.

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