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Neutron scattering measurements of magnetic excitations in Gd/Y superlattices

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Neutron inelastic scattering has been used to measure the magnetic excitations as a function of applied magnetic field in an antiferromagnetically coupled Gd/Y superlattice. The excitations were measured along the *c*-axis, which is parallel to the normal of the interfaces and the sample growth direction. Dispersive spin waves were unambiguously detected on the application of a magnetic field. The spin waves are shown to renormalize with field following a basic model drawn from standard spin wave theory. The model required no free parameters aside from an initial amplitude.

The development of thin film growth techniques has led to a revolution in electronics. Current research is focusing on spintronics,[1,2] whereby a device also exploits the spin of the electrons. These devices are inherently magnetic. Magnetic thin films and artificial structures also have an important role as model systems for the study of low dimensional magnetism[3] and magnetism in confinement.[4] Knowledge of the magnetic dynamics, and particularly spin waves, is critical to the understanding of thin film magnetism, from spin relaxation in devices[1] to the thermal stability of domain walls[2] to the competition between magnetic exchange and anisotropy.[3]

Appropriate experimental techniques are required to probe dynamics. A number of suitable techniques exist for magnetic thin films, although none of them are a panacea. Brillouin light scattering[5] and ferromagnetic resonance[6,7] are only able to probe dynamics close to a Brillouin zone center. Electron scattering and microscopy have recently been used to probe the magnetic dynamics of thin films. Spin-polarized electron energy loss spectroscopy has been used to measure magnons in a monolayer of iron,[8] but the technique has a very shallow depth penetration and is best suited for modes traveling on a surface. Inelastic scanning tunneling microscopy has been used on some transition metal crystals,[9,10] determining the energy of spin waves traveling normal to the surface by exploiting the formation of standing waves whose resonant frequencies change as a function of the film thickness.[10] Hence, the technique has limited access to the Brillouin zone of a single sample, and a dispersion curve can only be constructed by measuring many samples. Both techniques require careful sample preparation and control of the sample environment is technically complicated.

In principle, neutron inelastic scattering is the ideal technique for the study of magnetic dynamics. It is able to access the entire Brillouin zone in one sample, and has excellent penetration depth. It is also insensitive to sample environment and can be used in conjunction with high magnetic and electric fields, low temperatures, high pressures, etc. In comparison with other techniques, however, it suffers from low statistics, meaning that sample masses should be large, ideally a number of grams. This is obviously an issue when measuring thin films which contain considerably less than a gram of magnetic material.[11]

Nevertheless, experiments are feasible. This article reports the measurement of spin waves in a Gd/Y superlattice using inelastic neutron spectroscopy. The magnetic structure and properties of many rare-earth superlattices have been known for some time,[12] and this study is part of a broader effort which includes similar experiments on helimagnetic Dy/Y superlattices.[13] The spin waves were measured along the *c*-axis, which is normal to the interfaces and parallel to the sample growth direction. The data were measured in applied fields up to 5 T.

The sample measured for this study was a $\left[\mathrm{Y}_{17\ \text{Å}}/\mathrm{Gd}_{30\ \text{Å}}\right]_{70}$ superlattice with the crystallographic [0001] axes parallel to the surface normal, grown using molecular beam epitaxy methods on a 50×50 mm^2 substrate. The structure of the sample was confirmed with x-ray and neutron diffraction.

The neutron measurements were performed on the IN14 three-axis spectrometer at the Institut Laue-Langevin, France. The instrument was configured in a 'W' configuration with $40'$ collimation before and after the sample and a flat analyzer. The final wave vector was fixed to $k_f = 1.55$ Å^{-1} and the energy resolution at momentum transfer Q ~ 2.2 Å^{-1} was $\Delta E \approx 0.15$ meV.

Neutron diffraction data, measured at 5 K, are shown in Fig. 1. The nuclear structure peaks are separated by 0.130 ± 0.002 Å^{-1}, corresponding to a bilayer thickness

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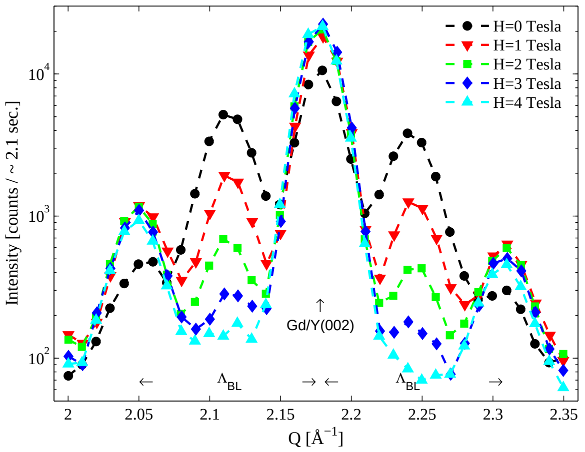

FIG. 1. Neutron diffraction data for the $\left[\mathrm{Y}_{17\ \text{Å}}/\mathrm{Gd}_{30\ \text{Å}}\right]_{70}$ superlattice as a function of applied external field. The measurements were taken at 5 K.

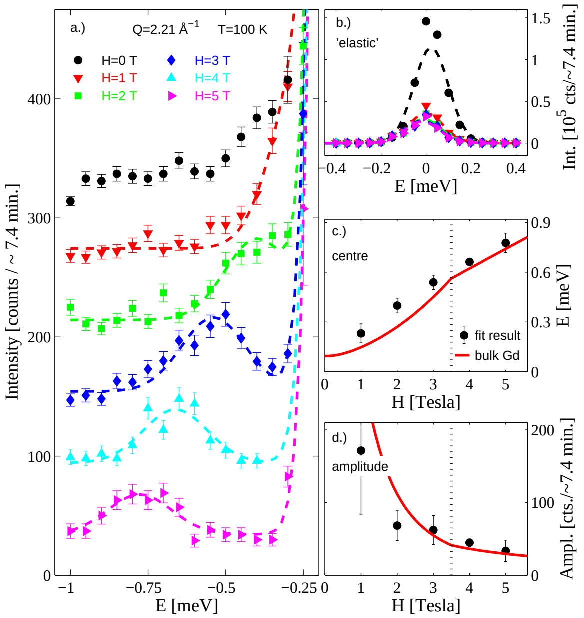

FIG. 2. Neutron inelastic data and analysis for the $\left[\mathrm{Y}_{17\ \text{Å}}/\mathrm{Gd}_{30\ \text{Å}}\right]_{70}$ superlattice. (a) Measurements of the inelastic signal at 100 K and Q = 2.21 Å^{-1} as a function of applied field. Data sets at increasing fields have been vertically shifted by 60 units relative to their predecessors. The dotted lines are fits of Gaussians to the data. Data at 5 K, representing an instrument background, are also shown. (b) Similar measurements on a different scale, incorporating the elastic data. (c) The characteristic energies of the inelastic peaks as a function of field. The solid line shows Eq. (2), and the critical field H_c is shown by the dotted line. (d) The fitted amplitudes to the inelastic peaks. The solid line shows Eq. (4) multiplied by an arbitrary amplitude of 18.

of $\Lambda_{\mathrm{BL}} = 48.3 \pm 0.7$ Å which is, within errors, consistent with the nominal thickness of the bilayer. Interface roughness was estimated at ~ 6 Å. The data at 0 T also show Bragg peaks at $\pi/\Lambda_{\mathrm{BL}}$ positions, indicating that the ferromagnetic Gd layers are antiferromagnetically coupled. While bulk Gd is ferromagnetic,[14,15] antiferromagnetic coupling has been observed and associated with the long-ranged nature of RKKY coupling across the nonmagnetic Y layers.[12] The half-order peaks disappear with increasing applied field as the Gd moments are forced along the field direction. At 4 T, all the Gd moments in the sample are ferromagnetically aligned.

The sample is estimated to have ~ 6 mg of magnetic material, which is very small for an inelastic neutron scattering experiment. To improve the chances of unambiguously observing spin waves, measurements were carried out at 100 K. Neutron diffraction measurements proved that the magnetic structure of the sample was the same at 5 and 100 K, and the neutron inelastic cross-section is significantly larger at elevated temperatures due to the thermal occupation, or “Bose”, factor. The spectra were measured in neutron energy loss. Due to the Bose factor, this part of the cross-section rapidly falls with increasing energy transfer at low temperatures. Thus, a reasonable estimate of the instrumental background could be determined by repeating a measurement at 5 K.

Initial measurements in zero field did not reveal any clear, dispersive inelastic signal. A signal, which could only be due to spin waves, did appear on the application of an external field. Figure 2(a) shows the data at Q = 2.21 Å^{-1}, and their evolution as the field is increased. The Q position is half way between the principal structural Bragg peak at Q = 2.178 Å^{-1}, a Brillouin zone center for the ferromagnetic structure corresponding to the mean (0002) Bragg peak position for Gd and Y, and the half order antiferromagnetic peak at Q = 2.25 Å^{-1}. The data could be comfortably fitted with two Gaussians: one for the elastic line shape shown in Fig. 2(b), and one for the spin wave that emerges as the field is increased. The fits are shown as dashed lines in Figs. 2(a) and (b). The widths for the spin wave Gaussians were fixed to $\Delta E_G = 0.24$ meV. This value was determined from the mean of the fit results at high fields where the spin wave is clearly resolvable. It is larger than the instrument resolution, but this is expected as the spin waves are dispersive and the total width of the signal results from a convolution of the resolution function with the dispersion surface. Fixing ΔE_G resulted in stable fits for the data at lower fields. The fitted centers and the amplitudes for the spin waves are shown in Figs. 2(c) and 2(d).

An apparent inelastic signal in 0 T was not fitted because, as shown in Fig. 2(b), the elastic signal is triple the intensity of those at larger fields. An elastic component may have entered in the resolution convolution, hence the apparent inelastic signal may be spurious. The same is not true for data measured in larger fields where the elastic signals are equivalent, hence these inelastic data may be self-consistently and reliably fitted.

The data may be described using a straight-forward model derived from the standard theory of spin waves,[16] incorporating the exchange parameters and the Zeeman

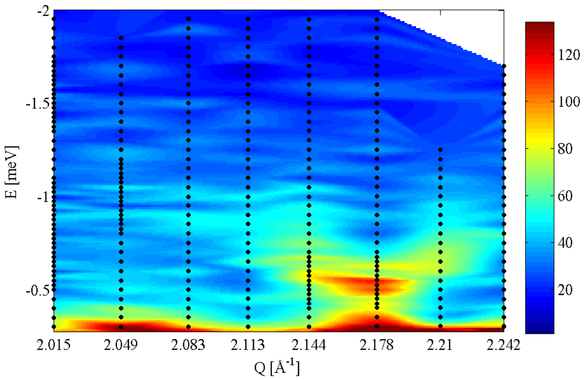

FIG. 3. Neutron inelastic scattering data measured at 100 K in an applied field of 4 T.

energy in an applied field. In zero field, this sample has antiferromagnetic coupling between the Gd layers. On the application of a small magnetic field, H, the moments will "flop" perpendicular to the field. Increasing the field will gradually force the moments along the field direction until a critical field, H_c, is reached, from which point all the moments will be collinear. From inspection of Fig. 1, the critical field may be estimated as $H_c \approx 3.5$ T.

In this model, the spin wave energies will renormalize with field following the equation:

$$\omega(q,H) = \sqrt{\omega_0(q,H)\left(\omega_0(q,H) + E_a\right)}, \tag{1}$$

where

$$\omega_0(q,H) = \omega_{ex}(q) + g\mu_B \left\{ \begin{array}{ll} SH^2/H_c, & 0 \le H \le H_c \\ SH, & H \ge H_c \end{array} \right. , \tag{2}$$

and

$$\omega_{ex}(q) = 2S\sum_{n=1}^{5} J_n \left(1 - \cos\frac{ncq}{2}\right). \tag{3}$$

Here, $S = 7$ is the spin on a Gd atom, c is the length of the crystallographic c-axis and J_n are the exchange constants. $\omega_{ex}(q)$ corresponds to the expression for spin waves as a function of reduced wavenumber, q, along the c-axis, and Eq. (3) is the function previously used to approximately model the spin waves in bulk Gd.[14,15]

The intensity of the inelastic signal is then given by:[17]

$$I(q,H) \propto \omega_0^{-1}(q,H) \tag{4}$$

A term, E_a, has been included to account for the easy-plane dipolar anisotropy known to be important for thin films.[12] The demagnetizing field has been estimated from saturation magnetization to be $4\pi M = 2.66$ T,[12] hence E_a was taken to be $E_a = g\mu_B 4\pi M = 0.3$ meV.

Thus, Eqs. (1) and (4) were used to model the data in Figs. 2(c) and 2(d), and the theory calculations have been included in the figures. The only free parameter in the comparison is a scaling constant for Eq. (4). Considering the simplicity of the model, the agreement is satisfactory.

Subsequent measurements at different Q along the [0001] direction are shown in Fig. 3. The measurements were made at 4 T, and clearly show a dispersive signal with a minimum at the mean (0002) Bragg peak position. The data are being analyzed in the framework of a more sophisticated model for the propagation of spin waves in superlattices and will be the subject of a future article.

In conclusion, neutron inelastic scattering has been used to measure spin waves in a Gd/Y superlattice and their evolution with the application of an applied field. The experiments, previously not considered to be feasible due to small sample masses, are possible and the data are of high quality. Neutron inelastic scattering is able to measure regions of the Brillouin Zone not accessible to other techniques, and the data in this report could not have been measured using any of the other known techniques. The analysis presented here is rudimentary but complete, with the model satisfactorily matching the data with effectively no free parameters. Future reports will use a more complete theory that has been developed for spin waves in superlattices to describe the scattering across the entire Brillouin zone. The theory will build on previous theories for spin waves in multilayers[18] but will be developed to account for the long-range magnetic interactions that are very important in the rare-earths.[15]

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