Magnetic Excitations of the Incommensurate Spin-Density Wave in Chromium Metal

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The magnetic excitations of the incommensurate spin-density wave in pure chromium have been ivestigated with use of inelastic neutron scattering. We have observed the spin wave modes originating from the incommensurate magnetic Bragg reflections. The polarizations of these modes are found to be highly anisotropic. We have also observed a new magnetic excitation centered at a commensurate wave vector. This new excitation is interpreted as a feature specific to a transverse incommensurate spin-density wave.

The discovery of the splitting of the magnetic Bragg reflections in chromium by Corliss, Hastings, and Weiss¹ about twenty years ago and the subsequent interpretation of this experiment in terms of itinerant spin-density wave (SDW) antiferromagnetism by Overhauser² and Lomer³ have created a long and continuing chapter in our understanding of magnetism in metals.

Although the magnetic excitations of chromium are of fundamental interest, these excitations of the incommensurate SDW have never been subject to investigation. Past studies of the spin waves in chromium^{4,5} used samples with a small addition of manganese which forces the modulation period to be commensurate with the bcc crystal lattice forming a simple Néel-type antiferromagnet. A very steep spin-wave dispersion and the availability of only small high-quality single crystals of pure chromium prevented past workers from investigating the excitations of the incommensurate state.

At temperatures below the spin-flip temperature ($T_F = 122$ K) the SDW is longitudinally polarized.⁶ Above T_F one finds the SDW in a state of transverse polarization up to the first-order transition at the Néel point $(T_N = 38.5 \text{ C}).^6$ In general, as a result of the cubic symmetry of chromium above $T_{\rm N}$ three types of domains develop below T_N with the wave vector \vec{Q} of the SDW along any one of the three [100]-type directions in the crystal giving rise to three sets of equivalent satellites. It is possible to force a single crystal of chromium to order into a single-Q domain by cooling through the Néel temperature in a large applied magnetic field.⁷ This procedure results in a crystal consisting of one type of domain with the modulation direction oriented along the applied field. The crystal will remain in such a

single- \overline{Q} state after removal of the field until being heated to a point above T_{N} .

The (100) reciprocal-lattice plane for a single- \vec{Q} crystal is shown in Fig. 1(b). The wave vector \vec{Q} is given by

$$\vec{\mathbf{Q}} = (2\pi/a)(0, 0, 0.9515)$$
 (1)

at low temperature and increasing slightly in magnitude with temperature.⁶ Here a is the lattice parameter. Magnetic Bragg reflections are observed at values of the neutron scattering vector

$$\vec{q} = \vec{G} \pm \vec{Q}, \tag{2}$$

where \vec{G} are the reciprocal-lattice vectors of the bcc lattice. The magnetization wave correspond-



FIG. 1. (a) Magnetization wave for the incommensurate SDW in pure chromium. (b) (100) Reciprocal-lattice plane of single- \vec{Q} chromium. The stars are the magnetic Bragg points.

ing to this modulation of the magnetic 3d electrons surrounding each atom by the SDW is shown in Fig. 1(a).

Both the past measurements^{4,5} and calculations^{8,9} indicate a very large spin-wave velocity in chromium, $\hbar C \simeq 1000$ meV Å. Such a high velocity presents special problems for typical triple-axis neutron spectroscopy because of the finite momentum resolution. A detailed discussion of these problems is presented by Als-Nielsen, Axe, and Shirane.⁵

In the experiments described here we have used a highly perfect single crystal of chromium of approximately 0.5 cm³ grown by vapor deposition in the reduction of chromium iodide. The crystal exhibited a narrow mosaic of less than 5' full width at half maximum (FWHM). A single- \bar{Q} state was induced by cooling through the Néel point in a magnetic field of 60 kOe aligned to within 0.5° of the [001] direction. The results of the field cooling were checked by measuring the intensity of the magnetic satellites around the (0, 0, 1) and (0, 1, 0) reciprocal-lattice positions. These measurements confirmed that more than 98% of the sample was in domains with \bar{Q} along the [001] direction.

The experiment was carried out on a triple-axis spectrometer at the Brookhaven National Laboratory high-flux-beam reactor with a fixed incident energy of 13.5 meV. In Fig. 2 the results of constant- ΔE scans along [001] across the satellites near the (0, 0, 1) and (0, 1, 0) points are presented. As expected, strong scattering is observed near the satellite positions where the spinwave dispersion surface is completely contained by the resolution ellipsoid. In spite of resolution effects, which complicate direct comparison of the two spectra, one can compare the peak intensities, because the orientation of the resolution ellipsoid with respect to the dispersion surface is equivalent for a momentum transfer of either $\vec{q} = (2\pi/a)(0, 0, 1 \pm \epsilon) \text{ or } \vec{q} = (2\pi/a)(0, 1, \pm \epsilon).$ After correcting the scan along \vec{Q} at $(0, 0, 1 \pm \epsilon)$ for the magnetic form factor, one can see that the scattering intensity at the $(0, 0, 1 \pm \epsilon)$ satellites is twice that of the $(0, 1, \pm \epsilon)$ satellites. Since the magnetic scattering is proportional to the magnetization fluctuations transverse to the momentum transfer¹⁰ q this result implies that the fluctuations of the SDW are confined to the x-y plane.

In addition to the spin-wave scattering in Fig. 2, there is clearly an additional feature centered at the commensurate point (0,0,1). A careful investigation around (0,1,0) reveals the presence



FIG. 2. Constant- ΔE scans across satellites in single- \vec{Q} direction. T = 200 K; $k_1 = 2.55$ Å⁻¹; collimation, 20'-40'-60'.

of this same feature at that point in the reciprocal lattice as well. The combined effects of a weaker intensity and a broadening of the spinwave scattering because of an instrumental resolution in a scan transverse \vec{q} render the peak unobservable in Fig. 2(b). In Fig. 3, results of con-



FIG. 3. Data taken for constant momentum transfer at the (0, 0, 1) position in reciprocal space.

stant-q scans at (0, 0, 1) for a series of temperatures are given. This more detailed examination of the scattering at (0, 0, 1) shows several interesting points. There is an excitation clearly centered at approximately 4 meV at T = 200 K. This peak in the scattering cross section grows slightly with increasing temperature and is centered at 3.5 meV at T = 300 K. The most dramatic change is in the very large sloping background that arises between 200 and 300 K. By 300 K the scattering for $\Delta E = -1.5$ meV has grown to over a factor of 30 above the level observed at T = 200 K. The increased cross section does not simply correspond to the increased spin-wave intensities between 200 and 300 K, as the spin-wave scattering has increased by only a factor of 2 in agreement with the change expected by the Boltzmann population factor. The background was also measured away from the (0, 0, 1) position; we found, for ΔE = -6.0 meV, only 50 counts/12 min at T = 290 Kcompared with 700 counts/12 min at (0, 0, 1). Constant-q energy scans at (0, 0, 1) and slightly above and below the spin-flip transition temperature clearly show that the 4-meV excitation is not present for the longitudinally polarized SDW. While the 4-meV peak can be clearly seen in the 130-K data, the 115-K data do not show even a suggestion of a peak in the scattering cross section. These data prove two important points: The central feature in the data of Fig. 2 is a real effect and not simply spin-wave intensity from the small amount of the crystal in a [100]-type domain. If the 4-meV excitation were somehow due to this type of spurious effect, the spin waves would still be visible below T_F with approximately the same intensity as above the spin-flip temperature. These data also show that the 4-meV excitation is somehow a feature of the transverse polarization state of the SDW.

The factor-of-2 difference in the spin-wave peak intensities in the two scans of Fig. 2 can be understood if the only allowed spin fluctuations are those transverse to \vec{Q} . In the longitudinal scan along [001] through the $(0, 0, 1 \pm \epsilon)$ satellites all fluctuations transverse to \vec{Q} contribute to the scattering cross section. However, in the scan along [001] through the $(1, 1, \pm \epsilon)$ satellites only half of the transverse fluctuations are perpendicular to \vec{q} and thus contribute to the scattering. This anisotropy hypothesis not only provides an explanation for the data of this experiment, but it is also consistent with other measurements on chromium. On the basis of their data, Werner, Arrott, and Kendrick⁶ concluded that there was a very large barrier to rotation of the spin polarization from transverse to longitudinal above T_F . Magnetic torque measurements¹¹ have demonstrated the anisotropy of the susceptibility in the transversely polarized SDW state.

The 4-meV excitation at (0, 0, 1) is a very interesting and important result of this experiment. suggesting that chromium has excitations which are specifically due to the incommensurate structure. Figure 1(a) shows that the amplitude of the spin density is reduced to a small value near the nodes of the SDW. For the case of only nearest neighbor coupling, each half period of modulated structure would be only weakly coupled to the adjacent regions. Although one must certainly include more than nearest-neighbor interactions in an itinerant antiferromagnet for a complete description, we will neglect the longer-ranged interactions for the purpose of developing a qualitative picture. A weak coupling between blocks of spins which are very strongly coupled internally immediately suggests the possibility of a low-energy excitation corresponding to large-amplitude fluctuations of the blocks of spins. Such a picture is very much like that of a domain wall or solitary wave where the large phase excursions occur in the region of weak interaction. It is extremely interesting to note that such excitations should be found at the commensurate point (0, 0, 1)with some natural width in momentum space due to the finite dimensions of such a block of spins. Below the spin-flip temperature, T_F , where the SDW is longitudinally polarized, one loses the degree of freedom associated with the transverse polarization. As we have shown, the excitation suddenly disappears at T_F consistent with this picture (see Fig. 4). In summary then, we believe that the commensurate excitation at $\Delta E = 4$ meV is due to the quantum mechanical fluctuation of the strongly coupled blocks of spins between the nodes of the modulated structure [(Fig. 1(a)] within the fourfold anisotropy potential in the x-yplane. We would interpret the large-sloping temperature-dependent scattering as due to excitations within and to the continuum of energy levels above the top of the anisotropy potential.

Future plans include experiments on the effects of a magnetic field on the 4.0-meV excitation in order to probe the role of anisotropy in the x-yplane. Other data from this experiment suggest the possibility of changes in the spin-wave spectrum below T_F . This problem is currently under investigation.

In conclusion, our studies of chromium have shown that above T_F the only allowed spin fluctuations are in the plane normal to the modulation direction. These experiments have also uncovered a new commensurate, low-lying, magnetic excitation of the incommensurate SDW.

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Influence of Nuclear Tracks on the Magnetic Properties of a (Gd, Bi)₃(Fe, Ga)₅ O₁₂ Garnet Film

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The temperature dependence of the saturation magnetization of an epitaxial $Gd_{2.3} Bi_{0.7}$ -Fe_{4.5} $Ga_{0.5} O_{12}$ garnet film irradiated with 180-MeV Xe ions of different doses has been investigated experimentally and analyzed theoretically in terms of molecular-field theory. The magnetic ordering inside the nuclear tracks turns out to be destroyed as a result of the large degree of disorder. The effective nuclear track diameter is estimated to be 15 nm. The molecular-field coefficients deduced from the magnetization curves depend on dose.

The effects induced by ion implantation in magnetic oxides have been studied intensively. Usually a thin layer of less than 1 μ m is implanted by light ions and with high doses of about 10^{16} cm⁻² giving rise to changes of the magnetic behavior. However, the influence on the magnetic properties of irradiation with heavy ions of high energy has not yet been reported except for some recent investigations on the relationship between the coercive force and the dose for Xe- and U-irradiated garnet films.¹⁻³ In contrast to ion implantation such ions induce nuclear tracks of cylindrical shape of $10-\mu m$ (180 MeV Xe) length. These tracks thus extend through the magnetic film of about 5 μ m into the substrate. In this work we study the effect of these tracks on the saturation magnetization of an epitaxial gadolinium iron garnet film.

A garnet film of composition $Gd_{2.3}Bi_{0.7}Fe_{4.5}$ - $Ga_{0.5}O_{12}$ epitaxially grown on a substituted gadolinium-gallium-garnet substrate has been irradiated with 180-MeV Xe ions (1.4 MeV/nucleon). This composition provides high Faraday rotation and thus permits also magneto-optic investigations. The sample of 2.5-cm diam was irradiated on $0.2-\times0.5$ -cm² areas with different doses. These areas were cut from the original sample and used for the measurements. All pieces had a small edge of nonirradiated materials whose area was determined with a microscope to be about 5% of the total area. The saturation magnetization was measured with a vibrating-sample magnetometer and the Faraday rotation with an optical hysteresisgraph at $\lambda = 633$ nm.

The temperature variation of M_s is displayed in Fig. 1, which is in accordance with that observed for the Faraday rotation.⁴ The data from the irradiated sections contain the contribution of the small nonirradiated edge. The corrected values are listed in Table I together with the Curie temperatures and the Faraday rotation. The M_s data of cut No. 4 for T > 420 K in particular