Polarization Dependence of the Dynamic Susceptibility $\chi''(\omega)$ in Chromium

J. E. Lorenzo, B. J. Sternlieb, and G. Shirane
Department of Physics, Brookhaven National Laboratory, Upton, New York 11973

S. A. Werner
Department of Physics, University of Missouri, Columbia, Missouri 65211
(Received 22 November 1993)

Neutron scattering measurements characterizing the polarization, energy, and temperature dependence of the magnetic excitations of the longitudinally polarized spin density wave phase of chromium are presented. Careful consideration of spectrometer resolution effects has resulted in the first quantitative expressions for the cross sections associated with the longitudinally and transversely polarized excitations. These results, together with a simple parametrization of the dynamic susceptibility, $\chi''(\omega) \propto A(\omega)/\omega$, reveal a simple dependence of these excitations on energy at all temperatures below the spin-flip transition.

PACS numbers: 75.25.+z

Chromium's role as the definitive spin density wave (SDW) system [1] has made it the focus of numerous experimental [2–6] and theoretical [7,8] studies over the years. At the Néel temperature, $T_N = 311$ K, paramagnetic chromium undergoes a transition to a transversely polarized spin density wave (TSDW) phase. The wave vector characterizing this SDW order, $\mathbf{Q}_\pm = (1 \pm \delta, 0, 0)$, reflects the nesting properties of hole and electron components of chromium's Fermi surface and is incommensurate with the underlying lattice. At the spin-flip temperature, $T_{sf} = 121$ K, a second transition to a longitudinally polarized spin density wave (LSDW) phase occurs. The single spin domain associated with a given magnetic satellite ($\mathbf{S} \parallel \mathbf{Q}_\pm$) in this phase allows a simple decomposition of the longitudinal and transverse excitation contributions to the overall scattering intensity. Below we present the results of measurements characterizing the polarization, energy, and temperature dependence of the excitations of the LSDW phase of chromium. Our focus on the dynamics of the LSDW phase is largely motivated by the unique role played by the longitudinal excitations as $\omega \to 0$ in this phase. No other magnetic system has evinced comparable excitations.

Efforts to measure the polarization dependence of the SDW excitations of chromium were first made by Fincher, Shirane, and Werner [2]. This work focused on the evolution of the longitudinal and transverse excitations through the spin-flip transition and at temperatures near T_N. At lower temperatures, the work of Burke *et al.* [3] demonstrated that longitudinal excitations dominate the low energy scattering in the LSDW phase. These measurements also demonstrated that the scattering intensities associated with the longitudinal and transverse excitations of the LSDW phase are strongly dependent on energy: Below $\Delta E \sim 9$ meV the longitudinal excitations are dominant while the scattering at higher energies is independent of polarization.

This Letter addresses two major concerns: First, no quantitative analysis has yet been performed to extract the cross sections, $\partial^2\sigma/\partial\omega\partial\Omega$, and corresponding dynamic susceptibilities, χ'', associated with the transverse and longitudinal excitations of the LSDW phase, and second, the temperature dependence of these modes has not yet been studied in any detail.

The study of these dynamics is complicated, below T_N, by the cubic symmetry of chromium which results in the formation of three magnetic domains. The scattering from these multiple domains is very difficult to resolve as the separation between the incommensurate magnetic satellites is comparable to practical vertical spectrometer resolutions. However, single domain or single-**Q** samples can be produced by cooling through T_N with a sufficiently strong magnetic field applied along the [100] crystal axis. The elastic magnetic peaks arising from this single domain are shown in the $(hk0)$ scattering plane in Fig. 1(a).

The polarizations of the LSDW excitations are readily determined by measuring the scattering intensities at both $(1 \pm \delta, 0, 0)$ and $(\pm\delta, 1, 0)$ as the cross section for the inelastic scattering of neutrons reflects only magnetic fluctuations normal to the neutron momentum transfer, $\boldsymbol{\kappa} \equiv \mathbf{k}_i - \mathbf{k}_f$. At $(1 \pm \delta, 0, 0)$, only fluctuations transverse to the LSDW polarization contribute to the scattering intensity, while the intensity measured at $(\pm\delta, 1, 0)$ reflects both longitudinal excitations parallel to the LSDW wave vector and transverse fluctuations perpendicular to the scattering plane of Fig. 1(a). Hence the contributions of the longitudinal and transverse fluctuations to the measured scattering intensities, neglecting variations in the magnetic form factor due to slightly different momentum transfers, can be expressed as

$$I_{(1\pm\delta,0,0)} = 2I_T, \quad I_{(\pm\delta,1,0)} = I_L + I_T. \tag{1}$$

Care must be taken in correcting the measured scattering intensities for the effects of instrument resolution. The high velocity, ~ 850 meV Å [9], of the SDW excita-

0031-9007/94/72(11)/1762(4)$06.00

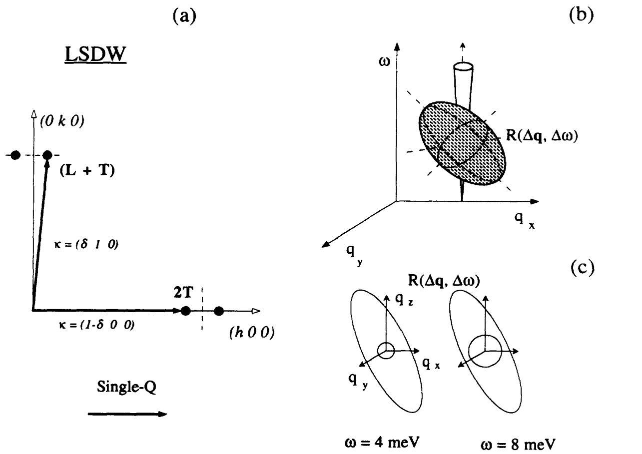

FIG. 1. (a) The $(hk0)$ scattering plane of chromium. Scans through $(1-\delta,0,0)$ measure only fluctuations transverse to the LSDW polarization $(2T)$, while the intensity measured at $(\delta,1,0)$ reflects both longitudinal excitations parallel to the LSDW wave vector and transverse fluctuations that are also orthogonal to the neutron momentum $(T+L)$. (b) The ellipsoidal neutron resolution function and the conelike excitation dispersion in ω-q space. (c) Resolution ellipsoids and spherical q shells which contribute to the observed scattering intensity. The ellipsoidal surface of the resolution volume represents the Gaussian falloff of the scattering sensitivity to half of its peak value.

tions in chromium results in a very steep, resolution limited "cone" of scattering, emanating from elastic magnetic satellite positions, as shown in the ω-q diagram, Fig. 1(b). The narrow momentum width of this dispersion surface relative to the ellipsoidal resolution function results in a peak scattering intensity, centered at the magnetic satellite position, that reflects the entire range of q values with excitation energies within the energy resolution of the spectrometer. These q values can be represented by a spherical shell in q^3 space as shown in Fig. 1(c). The actual measured peak intensities are proportional to the surface area of this shell, $\sim q^2$. As shown by Sinha *et al.* [9], the analysis of scattering data can be simplified by incorporating this dependence explicitly in the formulation of the scattering intensity. Assuming a linear dispersion relation [10], $\omega_q \propto q$, we have

$$I_{T,L} = \langle n(\omega_q)+1\rangle \frac{A_{T,L}(\omega_q)}{\omega_q}\omega_q^2 \mathcal{C}(\omega_q)\,, \qquad (2)$$

with

$$\chi''_{T,L}(\omega) = \frac{A_{T,L}(\omega)}{\omega}\delta(\omega-\omega_q)\,. \qquad (3)$$

Here ω_q^2 is the approximation term mentioned above and $n(\omega_q)$ is the boson occupation number. This ω_q^2 approximation is valid at low energies; however, a significant correction, $\mathcal{C}(\omega_q)$, must be applied at higher energies as the size of the shell is not negligible compared to the q^3-space extent of the resolution ellipsoid as shown in Fig. 1(c). For the spectrometer configuration used in this experiment, this term amounts to $\mathcal{C}(4\text{ meV}) = 0.82$ and $\mathcal{C}(8\text{ meV}) = 0.63$. We note that the $1/\omega$ parametrization of the dynamic susceptibility, Eq. (3), is the same as used in the analysis of antiferromagnets with localized moments. In the case of traditional antiferromagnetic spin waves, the parameter $A_T(\omega)$ is a constant and $A_L(\omega) = 0$.

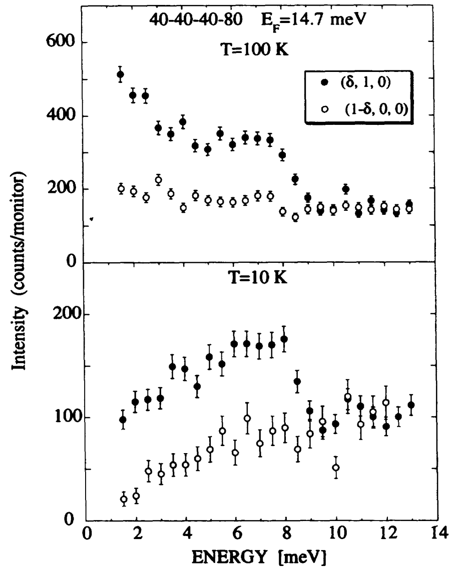

FIG. 2. $(1-\delta,0,0)$ and $(\delta,1,0)$ peak intensity data at $T = 100$ and 10 K.

In Fig. 2 we show $(1-\delta,0,0)$ and $(\delta,1,0)$ peak intensi-

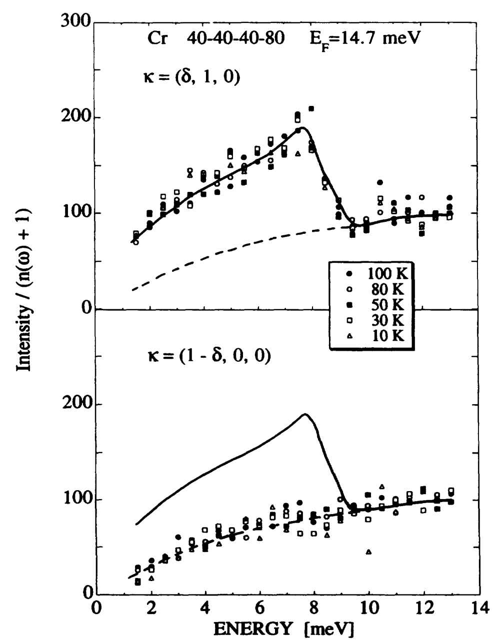

FIG. 3. Thermally corrected $(1-\delta,0,0)$ and $(\delta,1,0)$ peak intensity data, $I/\langle n(\omega)+1\rangle$, at $T = 100$, 80, 50, 30, and 10 K.

ty data at $T=100$ and 10 K. These measurements are in qualitative agreement with the original $T=55$ K results of Burke *et al.* [3]. As argued in Eq. (1), the equality of the scattering from these two satellites above $\Delta E \sim 9$ meV implies that the intensities of the longitudinal and transverse fluctuations in this energy range are equal. At lower energies the intensity associated with the $(\delta,1,0)$ satellite increases abruptly and peaks, in the 10 K data, at $\Delta E \sim 8$ meV. The smooth energy dependence of the purely transverse, $(1-\delta,0,0)$, data argues that this dramatic behavior is entirely due to the energy dependence of the longitudinally polarized excitations.

To study the dependence of the dynamic susceptibility, χ'', on temperature, the thermal factor of Eq. (2), $\langle n(\omega)+1\rangle$, was removed from the 10 and 100 K data of Fig. 2 and additional, constant-**Q** scans at 30, 50, and 80 K. The resulting $(1-\delta,0,0)$ and $(\delta,1,0)$ peak intensities are plotted in Fig. 3. These data clearly show that the susceptibilities associated with both the longitudinal and transverse fluctuations are independent of temperature. In particular, the $\Delta E=8$ meV energy scale defined by the energy dependence of the longitudinal excitations is a constant characteristic of the LSDW phase of pure chromium.

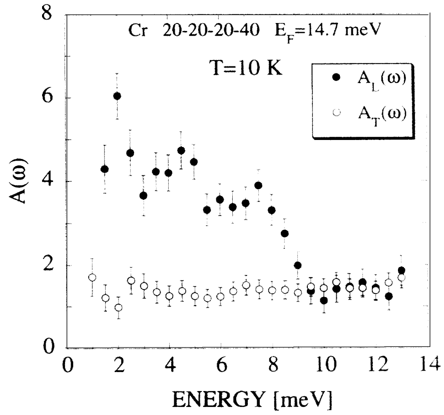

FIG. 4. The parameters $A_{T,L}(\omega)$ which characterize the dynamic response. The constant character of $A_T(\omega)$ indicates that the transverse LSDW excitations have a simple $\chi''_T(\omega) \sim 1/\omega$ dependence at all energies below $\Delta E \le 14$ meV.

Finally, Eqs. (1)–(3) can be inverted to extract resolution corrected longitudinal and transverse susceptibility parameters as follows:

$$A_{T,L}(\omega_q)=\frac{1}{\omega_q}\frac{1}{\mathcal{C}(\omega_q)}\frac{1}{\langle n(\omega_q)+1\rangle}\times\begin{cases} I_{(1\pm\delta,0,0)}-\frac{1}{2}I_{(\pm\delta,1,0)} & (L)\\ \frac{1}{2}I_{(\pm\delta,1,0)} & (T)\end{cases}$$

The parameters $A_{T,L}(\omega)$, which characterize the dynamic response, are plotted in Fig. 4. The constant character of $A_T(\omega)$ indicates that the transverse LSDW excitations have a simple $\chi''_T(\omega)\sim 1/\omega$ dependence at all energies below $\Delta E \le 14$ meV. As mentioned above, the spin wave excitations of conventional antiferromagnets evince the same behavior. This similarity is surprising as the sinusoidal spin modulation of chromium is physically quite different from the constant, fixed moment character of these systems. In contrast, the steplike behavior of $A_L(\omega)$ has no magnetic analog. This sharp feature, and the agreement of $\chi''_L(\omega)$ and $\chi''_T(\omega)$ at higher energies, suggest that the longitudinal susceptibility may be the sum of terms corresponding to different, distinct underlying excitation mechanisms. Recent additional data also suggest that, at low energies, the energy dependence of $A_L(\omega)$ may have structure in addition to the peak at $\Delta E=8$ meV.

We note that the analysis presented in this Letter has a nontrivial dependence, via $\mathcal{C}(\omega)$, on the neutron resolution function. A forthcoming paper will address this issue in greater detail. However, we mention that the incorporation of resolution corrections has been checked by varying both spectrometer energy and collimation configurations. For instance, the $T=10$ K measurements of Fig. 2, taken with a 40′-40′-40′-80′ collimator configuration and a fixed, final energy of 14.7 meV, lead to results that are consistent with the 20′-20′-20′-40′ data presented in Fig. 4.

Our results stress the importance of characterizing the energy dependence of $\chi''(\omega)$ in terms of the parameter $A(\omega)$. As shown in Fig. 4, the removal of the $1/\omega$ dependence from $\chi''(\omega)$ dramatically demonstrates the contrast between the flat, conventional character of $A_T(\omega)$ and the more exotic behavior of $A_L(\omega)$.

We would like to thank E. Fawcett, S. K. Sinha, R. S. Fishman, and S. H. Liu for many stimulating discussions. Invaluable technical assistance was provided by R. Rothe, J. Biancarosa, and R. J. Liegel. Work at BNL was carried out under Contract No. DE-AC0276CH00016, Division of Materials Science, U.S. Department of Energy. S.A.W. would like to acknowledge the support of the NSF through Grant No. NSF-PHY 9024608.

[1] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).
[2] C. R. Fincher, G. Shirane, and S. A. Werner, Phys. Rev. Lett. **43**, 1441 (1979).
[3] S. K. Burke, W. G. Stirling, K. R. A. Ziebeck, and J. G. Booth, Phys. Rev. Lett. **51**, 494 (1983).
[4] B. J. Sternlieb, G. Shirane, S. A. Werner, and E. Fawcett, Phys. Rev. B **48**, 10217 (1993).
[5] R. Pynn, W. G. Stirling, and A. Severing, Physica **180 & 181B**, 203 (1992).

[6] S. A. Werner, E. Fawcett, M. W. Elmiger, and G. Shirane, J. Appl. Phys. (to be published).
[7] A. Shibatani, K. Motizuki, and T. Nagamiya, Phys. Rev. **177**, 984 (1969).
[8] R. S. Fishman and R. H. Liu, Phys. Rev. B **48**, 3820 (1993).
[9] S. K. Sinha *et al.*, Phys. Rev. B **15**, 1415 (1977).
[10] Measurements have not demonstrated that these fluctuations follow a linear dispersion. However, the measurements of Sinha *et al.* [9] on CrMn suggest that this assumption can reasonably be extended to the excitations of pure chromium.

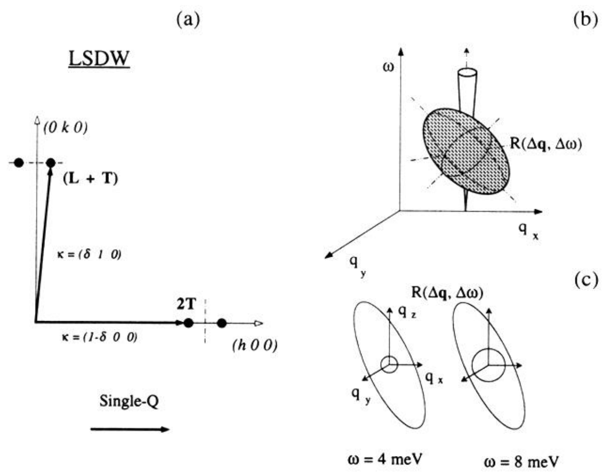

FIG. 1. (a) The $(hk0)$ scattering plane of chromium. Scans through $(1-\delta,0,0)$ measure only fluctuations transverse to the LSDW polarization $(2T)$, while the intensity measured at $(\delta,1,0)$ reflects both longitudinal excitations parallel to the LSDW wave vector and transverse fluctuations that are also orthogonal to the neutron momentum $(T+L)$. (b) The ellipsoidal neutron resolution function and the conelike excitation dispersion in ω-q space. (c) Resolution ellipsoids and spherical q shells which contribute to the observed scattering intensity. The ellipsoidal surface of the resolution volume represents the Gaussian falloff of the scattering sensitivity to half of its peak value.