Magnetic Fluctuation Scattering from Metallic Nickel above the Curie Temperature

O. Steinsvoll,[a] C. F. Majkrzak, G. Shirane, and J. Wicksted
Brookhaven National Laboratory, Upton, New York 11973
(Received 17 May 1983)

The spectral distribution of the magnetic fluctuation scattering from metallic nickel has been studied above the Curie temperature with unpolarized and polarized neutrons. The present measurements confirm previous results of a spin-wave-like mode above T_c. It is shown however, that the main part of the magnetic scattering above T_c does not come from the spin-wave ridge in $(\omega, \vec{q})$ space but from a spectral part centered at zero energy with a broad energy distribution.

PACS numbers: 72.25.+z, 75.50.Cc

The most remarkable property of metallic nickel is its ability to sustain propagating collective modes of spin-wave character well above the Curie temperature T_c.[1,2] Similar features have also been observed in metallic iron[3] and in the intermetallic compound MnSi.[4] Recently, a series of scattering experiments at the Institute Laue-Langevin, Grenoble, have been reported utilizing polarized neutrons and polarization analysis on 3*d* transition metals and compounds in the paramagnetic state [MnSi, Cr, Fe(Si)].[5-7] In these experiments the Grenoble group has utilized, intentionally, a very poor energy resolution (~ 43 meV) in order to obtain a proper value for $S(\vec{Q})$, the integral over ω of the scattering function $S(\vec{Q}, \omega)$. The results of the latter experiments have been interpreted as showing a considerable short-range order which persists up to many times T_c.

Ishikawa[8] recently presented the argument that, in the case of MnSi, the spin waves above T_c previously reported would be sufficient to explain the new Grenoble results. If this is true, then a similar explanation may also be valid for the other metals investigated by the Grenoble group. Because of the poor resolution, the Grenoble group had difficulties in separating the scattering into elastic and inelastic parts. It is, therefore, of utmost importance to characterize $S(\vec{Q}, \omega)$ properly for metallic Fe and Ni in order to resolve how much the spin waves contribute to the magnetic fluctuation scattering above T_c. We have examined Ni first and a subsequent study of Fe is now being undertaken. Here, we give a preliminary report of our experiments on nickel using both unpolarized and polarized neutrons with an energy resolution sufficient to characterize the spectral shape of the magnetic fluctuation scattering above T_c. Our measurements overlap previous experiments on Ni,[9] for small values of q in the critical temperature region. For larger q values our measurements extend into the region where a spin-wave ridge has been observed.[1,10]

Inelastic neutron-scattering experiments were performed on the same cylindrical single crystal of ^{60}Ni as was used by Minkiewicz *et al.*,[9] which has a volume of 2.9 cm^3. With this particular isotope the nuclear spin and isotopic incoherent scattering are eliminated and the phonon scattering is reduced by an order of magnitude relative to natural nickel because of the small nuclear scattering amplitude of ^{60}Ni. The $[0\bar{1}1]$ axis was chosen to be perpendicular to the scattering plane. The data were collected in the vicinity of (111) by triple-axis scans in the creation mode in the [111] direction. When unpolarized neutrons were used, a bent pyrolytic graphite analyzer was fixed at 14.5, 30.5, or 60 meV with all collimators at 20′, thus giving an energy resolution (full width at half maximum) of 0.5, 1.4, and 4 meV, respectively. The most useful data were collected at 30.5 meV. In the polarized-neutron case we used vertically magnetized composite Heusler-alloy monochromators and analyzers with magnetic guide fields and a flat-coil spin flipper in the beam path. In the latter case the focusing analyzer was fixed at 30.5 meV and all collimators were 40′ giving an energy resolution of 2.8 meV. The guide field at the sample was along the scattering vector and therefore horizontal. In this geometry the paramagnetic scattering is spin flip.[11] Since there is no nuclear-spin incoherent scattering from the sample, the depolarization in the sample is small, and the instrumental flipping ratio is high ($R = 18$); practically all of the neutrons collected with the flipper on are therefore due to magnetic scattering, with either paramagnetic or spin-wave origin.

The solid and dotted lines in Fig. 1(a) show the spin-wave (SW) dispersion relation at two different temperatures below and above T_c as obtained by Mook and co-workers.[1,2] The lower line is

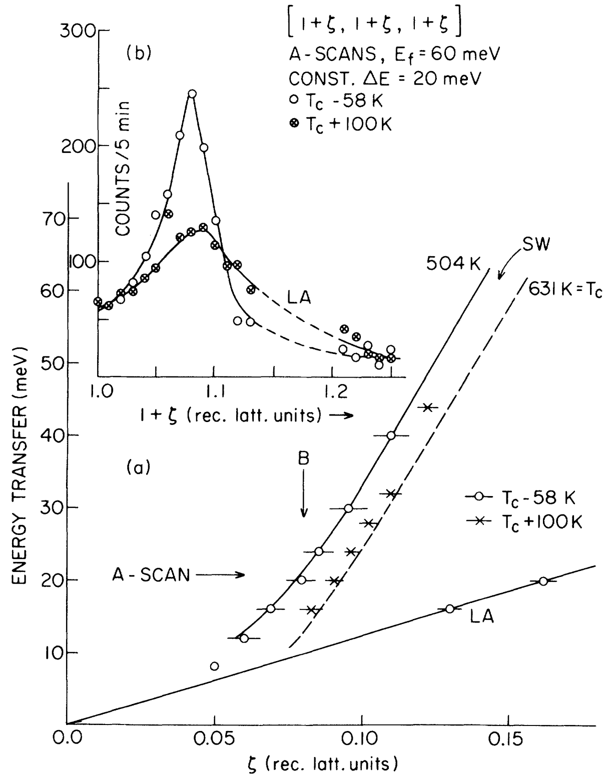

FIG. 1. (a) The spin-wave (SW) dispersion curves for two temperatures, the full line for 504 K and the broken line for 631 K ($=T_c$) taken from Ref. 2. The circles and the crosses are positions of spin-wave peaks at T_c -58 K and T_c+100 K as obtained by us. The longitudinal acoustic phonon branch is labeled LA. (b) Spin-wave peaks measured with a constant energy transfer of 20 meV below and above T_c (*A* scans). The curves are guides to the eye.

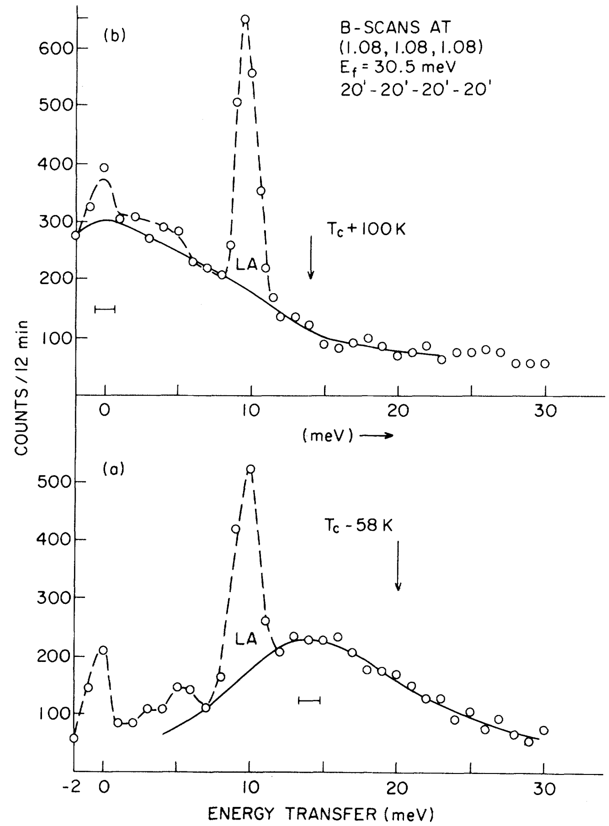

FIG. 2. The results of scans at constant momentum transfer at (1.08, 1.08, 1.08) for T_c-58 K and T_c+100 K (*B* scans). The approximate shape of the magnetic fluctuation scattering has been drawn with full lines. In between the LA peak and a small elastic scattering peak, a small amount of transverse phonon scattering is observed. Both full and broken lines are guides to the eye. The arrows point towards the expected spin-wave peak. The horizontal bars show the resolution.

the dispersion curve[12] for longitudinal acoustic (LA) phonons. Our spin-wave peak positions obtained by constant-energy scans (*A* scans) with unpolarized neutrons are shown as circles and crosses in Fig. 1(a). These scans were taken at 58 K below and 100 K above T_c. The peak positions show very close agreement with the measurements in Ref. 2, thus confirming the presence of a persistent spin-wave mode. Examples of such constant-energy scans are shown in Fig. 1(b) for an energy transfer of 20 meV. The LA-phonon peaks have been deleted from the plot to show more clearly the pileup of intensity across the spin-wave ridge.

In Figs. 2(a) and 2(b) we have plotted two typical scans for constant momentum transfer, i.e., scans labeled *B* in Fig. 1(a). The approximate distribution of the magnetic fluctuation scattering has been drawn with heavy lines in the plot. Superimposed on the magnetic ridge we see background elastic scattering, the LA phonon peak, and in between some scattered intensity due to interference by the transverse phonon branch. From Fig. 2(a), at 58 K below T_c, we see that the maximum of magnetic scattering is shifted to a lower energy value than expected from the dispersion curve in Fig. 1(a). This can be explained by the combined effects of the resolution function and the very steep slope of the spin-wave dispersion relation. The plot in Fig. 2(b), at 100 K above T_c, shows that the distribution of magnetic scattering is now centered around zero energy transfer. The arrow in Fig. 2(b) points towards the expected spin-wave peak position. No spin-wave peak can be seen in this constant-Q scan. Our measurements, therefore, show that even in

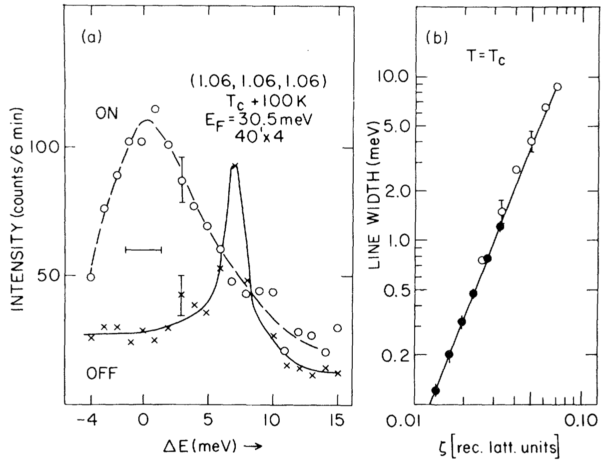

FIG. 3. (a) The points along the broken line ("ON") show a constant-Q scan with polarized neutrons and spin-flip analysis for $T_c + 100$ K. The crosses along the full line ("OFF") show the same scan but with non-spin-flip analysis. The polarization analysis effectively separates paramagnetic scattering from phonon scattering. The lines are guides to the eye. (b) The open circles show experimental half widths at half maximum obtained by us at T_c for $E_F = 14.7$ meV. The filled circles are half widths at small q from Ref. 9. The straight line is the best fit to these data.

the region of q where spin waves exist above T_c, the dominating contribution to the magnetic scattering comes from the regions around zero energy transfer. A similar constant-Q scan using polarized neutrons and spin-flip polarization analysis at 100 K above T_c is shown in Fig. 3(a). The result is the same. As the magnetic scattering and phonon scattering are well separated by this technique, the smooth tail of the ON curve [open circles in Fig. 3(a)] indicates even more convincingly that no peak exists at the expected spin-wave position. Both types of measurements are also consistent in showing no strong elastic contribution in the Q region covered in our experiments. From the constant-Q measurements at 50 and 100 K above T_c we find that the integrated magnetic intensities for momentum values larger than (1.06, 1.06, 1.06) reciprocal lattice units change very little with temperature.

The open circles in Fig. 3(b) show the half widths at half maximum of the magnetic scattering as obtained by us at T_c. The straight line has been drawn on the basis of the small-q data.[9] Our widths have not been corrected for resolution effects but still fit in nicely as an extension of the small-q data at T_c.

The overall picture described above for metallic nickel lends support to some of the conclusions drawn by the Grenoble group about the magnetic scattering observed above T_c from the compound Fe(Si). Our results agree with theirs in that for both nickel and iron the intensity due to spin-wave scattering is very much less than that due to paramagnetic scattering. Also, the temperature dependence at different Q values is similar in the two metals. We have further demonstrated that the paramagnetic scattering from nickel has a considerable energy width and that there is no separate purely elastic contribution.

In all their papers the Grenoble group has been able to normalize their scattered intensities so as to obtain absolute cross-section values. This is a valuable contribution which leads the trend towards more quantitative statements about magnetic neutron scattering. In a forthcoming paper we will follow up this trend and assign absolute cross-section values to our experimental intensities.

During the course of our experiments, we have learned that Lynn[13] has recently been conducting experiments on nickel above T_c for high Q values along the magnetic ridge, i.e., at and above (1.2, 1.2, 1.2) reciprocal lattice units. This is, however, outside the momentum-transfer range of our study. In addition we have recently become aware of the latest Grenoble paper[14] where they describe results of experiments on natural polycrystalline metallic nickel around (000) at 700 K. After correction for the magnetic form factor and conversion of our data around (111) to absolute cross sections there is reasonable agreement between our data obtained at $T_c + 50$ K and the Grenoble results.

This work was supported in part by the Division of Materials Sciences, U. S. Department of Energy under Contract No. DE-AC02-76CH00016.

[a]On leave from Institute for Energy Technology, 2007 Kjeller, Norway.

[1]H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).
[2]J. W. Lynn and H. A. Mook, Phys. Rev. B 23, 198 (1981).
[3]J. W. Lynn, Phys. Rev. B 11, 2624 (1975).
[4]Y. Ishikawa, G. Shirane, J. A. Tarvin, and M. Kohgi, Phys. Rev. B 16, 4956 (1977).
[5]K. R. A. Ziebeck and P. J. Brown, J. Phys. F. 10, 2015 (1980).
[6]K. R. A. Ziebeck, J. G. Booth, R. J. Brown, H. Ca-

pellmann, and J. A. C. Bland, Z. Phys. B 48, 233 (1982).

[7]P. J. Brown, H. Capellmann, J. Déportes, D. Givord, and K. R. A. Ziebeck, J. Magn. Magn. Mater. 30, 243 (1982).

[8]Y. Ishikawa, in Proceedings of the International Conference on Magnetic Materials, Kyoto, 1982 (to be published).

[9]V. J. Minkiewicz, M. F. Collins, R. Nathans, and G. Shirane, Phys. Rev. 182, 624 (1969).

[10]H. A. Mook and D. Tocchetti, Phys. Rev. Lett. 43, 2029 (1979).

[11]R. M. Moon, T. Riste, and W. C. Koehler, Phys. Rev. 181, 920 (1969).

[12]R. J. Birgeneau, J. Cordes, G. Dolling, and A. D. B. Woods, Phys. Rev. 136, A1359 (1964).

[13]J. W. Lynn, private communication.

[14]P. J. Brown, H. Capellmann, J. Déportes, D. Givord, and K. R. A. Ziebeck, J. Magn. Magn. Mater. 31-34, 295 (1983).