Spiral magnetic correlation in cubic MnSi

G. Shirane, R. Cowley,* and C. Majkrzak
Brookhaven National Laboratory, Upton, New York 11973

J. B. Sokoloff, B. Pagonis, and C. H. Perry
Northeastern University, Boston, Massachusetts 02115

Y. Ishikawa
Tohoku University, Sendai 980, Japan
(Received 11 July 1983)

MnSi is a cubic ferromagnet with a long-wavelength spiral below its Curie temperature at 29 K. Recent theoretical and experimental studies have shown that this spiral is only right handed because of the lack of a center of symmetry in the Mn atomic arrangement. We have carried out elastic and inelastic neutron scattering experiments on a MnSi single crystal with the use of a neutron polarized-beam technique. A characteristic polarization dependence was observed in the magnetic Bragg reflections of the spiral phase, as well as the spin-wave excitations and the critical scattering near T_c. This polarization dependence persists into the ferromagnetic phase under a magnetic field and also at temperatures as high as 150 K.

I. INTRODUCTION

MnSi is a unique itinerant-electron ferromagnet with a Curie temperature T_c of 29 K and an ordered magnetic moment of $0.4\mu_B$ on each Mn atom. The magnetic structure in the absence of a field is a long-period ferromagnetic spiral[1] with the propagation vector $(2\pi/a)(\zeta,\zeta,\zeta)$ with $\zeta=0.017$, but as shown in the phase diagram in Fig. 1 the magnetic structure becomes ferromagnetic in fields larger than 6 kOe. The magnetic neutron scattering of MnSi has been extensively investigated in recent years[2,3]; the most remarkable characteristic is the Stoner-type excitation[2] which persists up to temperature $T\approx 10T_c$. A more recent study[4] by Ziebeck *et al.* demonstrated that the total magnetic scattering cross section (integrated over energy) also shows strong short-range correlations at high temperatures.

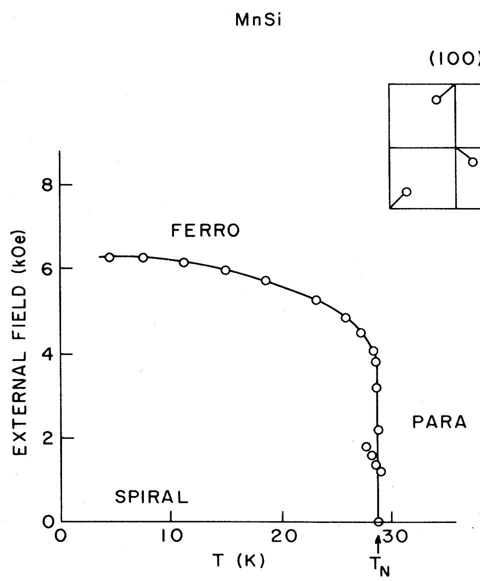

FIG. 1. Magnetic phase diagram of MnSi. Inset shows the projection of Mn atomic positions on a (100) plane. This atomic arrangement lacks a center of symmetry.

Recently, MnSi attracted attention from an entirely different angle. Theoretical work by Bak and Jensen[5] and Nakanishi *et al.*,[6] have shown that the ferromagnetic spiral in MnSi is caused by a Dzyaloshinski interaction which arises because of the noncentral arrangement of the Mn magnetic atoms in the unit cell (Fig. 1). The magnetic spiral in MnSi is then predicted to be different from those arising from competing exchange interactions, as discussed by Yoshimori,[7] because the spiral in MnSi must be only right (or left) handed, depending upon the sign of the Dzyaloshinski term in the Hamiltonian. This theoretical prediction can be tested by experiments using polarized neutrons because a single-handed spiral scatters only a particular polarization of the neutrons. This experiment was performed recently[8] and established that the spiral is right handed. Note that in the regular spiral structures, both right- and left-handed spirals coexist with equal probabilities, so that the scattering gives equal amount of both polarizations of the neutrons.

We came across this one-handed spiral problem accidentally while we were engaged in the study of forbidden magnons[9,10] in MnSi. We were aware of the theoretical[5,6] and experimental[8] work on the one-handed spiral of MnSi but we did not expect this property would effect measurements in the ferromagnetic phase at 7.5 kOe and at high temperatures. Surprisingly, we found that the one-handed spiral does have a very strong influence on the scattering in all regions in the H-T phase diagram shown in Fig. 1.

The main purpose of the present paper is to discuss the nature of the diffuse magnetic scattering and the inelastic scattering cross section. However, we first characterize our sample by describing the field-cooling effect on the magnetic Bragg scattering. The first observation of the right-handed spiral for MnSi is planned to be reported by Ishida *et al.*[8]

Before we describe our experimental arrangement, it is worthwhile to describe the arrangement of the Mn atoms in MnSi. The space group is the cubic group $P2_13$ (T^4) with lattice parameter $a = 4.558$ Å. The four Mn atoms are situated at (u,u,u), $(\frac{1}{2}+u,\frac{1}{2}-u,-u)$, $(\frac{1}{2}-u,-u,\frac{1}{2}+u)$, and $(-u,\frac{1}{2}+u,\frac{1}{2}-u)$ with $u = 0.138$. The projection of these Mn atoms on the (001) plane is shown in Fig. 1. This cubic structure not only lacks inversion symmetry but it also possesses the unusual characteristic that the intensity of a (210) reflection is different from that of (120). We emphasize that the chemical atomic arrangement does not have a spiral nature but it does lack a center of symmetry. Theoretically,[5,6] this is sufficient to create a single-handed spiral and the chirality (right or left handed) is uniquely determined by the sign of the Dzyaloshinski term in the Hamiltonian.

II. EXPERIMENTAL ARRANGEMENT AND BRAGG SCATTERING

The single crystal used in this study was the same sample as that used in earlier measurements.[2] It is a large cylindrical sample 12 mm in diameter and with a [110] axis nearly parallel to the cylinder axis. All the measurements were performed with this [110] axis vertical.

The experimental arrangements used in the polarized-beam experiments are shown schematically in Fig. 2. Heusler crystals were used as polarizers, and since the magnetic structure factor for this crystal's (111) reflection is negative, it reflects neutrons which are polarized antiparallel (−) to the field applied to the Heusler crystal, which was vertical with the *N* pole up. If the flipper is turned on (ON in figure), then the crystal reflects the neutrons which are polarized parallel (+) at the specimen. Other polarizers such as CoFe and multilayers all reflect neutrons with a polarization parallel to the applied field and so reflect (+) neutrons with the flipper off (OFF in figure).

When the horizontal field $\vec{H}$ is applied to the sample along the scattering vector, $\vec{Q} = \vec{k} - \vec{k}'$, then all the magnetic scattering—Bragg, paramagnetic, or spin wave—gives rise to a flip of the neutron spin.[11] In paramagnetic or antiferromagnetic materials, full polarization analysis, setup III of Fig. 2, is needed to separate out the magnetic from the nuclear scattering. In ferromagnetic crystals, the "half"-polarized arrangements (setups I and II) can be used to separate the spin-wave scattering as shown in Fig. 2(b). Both of these arrangements are equivalent except that setup II (in which the Heusler crystals is a monochromator) is less efficient than setup I.[12]

The magnetic Bragg scattering in the spiral phase of MnSi was studied using arrangement I. The crystal was cooled in a magnetic field with the field aligned along the [111] direction. This procedure eliminates the domains with the spirals having wave vectors *C* and *D* in Fig. 3 and when fields between 0.5 and 10 kOe were used, the amount of domain *C* was only about 1% of that of domain *A*. The scattering was measured as a function of the applied field between 100 Oe and 100 kOe. Experiment showed that the domain boundaries in the spiral of MnSi caused little depolarization of the beam as the flipping ratio of the polarized neutrons varied between 100 and 20.

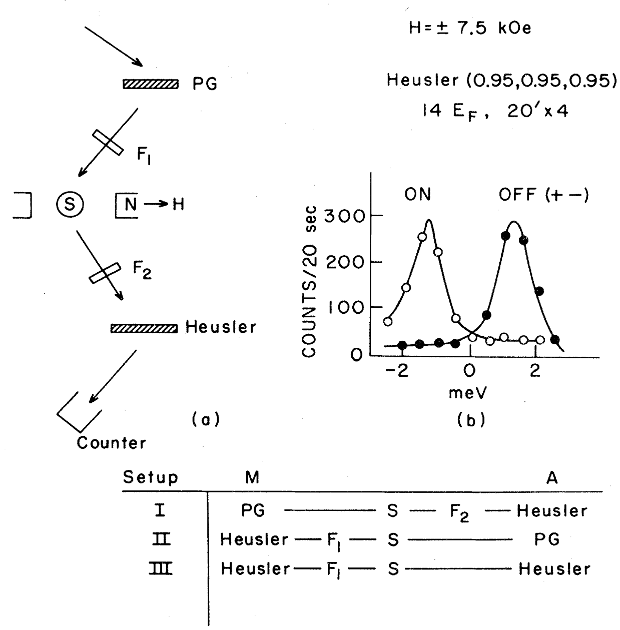

FIG. 2. Three experimental setups for polarized-beam experiments are shown in the inset table. Most experiments are done with setup I, shown in (a) with examples of polarization dependence of Heusler crystal sample in (b).

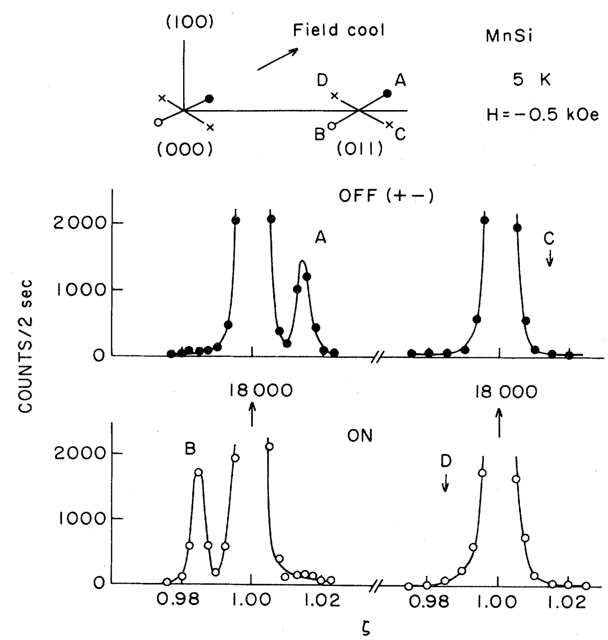

FIG. 3. MnSi crystal was field cooled along the [111] direction. Domains *C* and *D* are eliminated and *A* and *B* show characteristic polarization dependence of a one-handed spiral.

The polarization dependence of the scattering by a spiral structure was discussed by Blume,[13] who showed that a one-handed spiral would scatter only one polarization state of the neutrons. The experimental results are shown in Fig. 3 and they clearly show that the A and B satellites scatter the neutrons with different states of polarization. The sense of the magnetic field parallel to $\vec{Q}$, Fig. 2(a), shows that each of these satellites correspond to a right-handed spiral. When the horizontal magnetic field is reversed to -0.5 kOe the scattering from the A satellite occurs with the flipper on showing that the spiral remains in the same sense when the field is reversed.

We note that the A and B satellite scattering does *not* come from the same part of the crystal but corresponds to different domains. In principle, the relative population can be unbalanced by applying a mechanical stress or by an internal strain. We have tried to demonstrate that the A and B scattering are not coming from the same area by taking polaroid pictures of the scattered neutrons at the two positions. They show definitely different shapes. However, careful examination indicates that this is created by a nonuniformity of mosaic within the crystal. When the crystal is realigned at A and B positions to give maximum scattering, the two pictures become very similar. Maybe a small and more perfect crystal might reveal the different A and B domains.

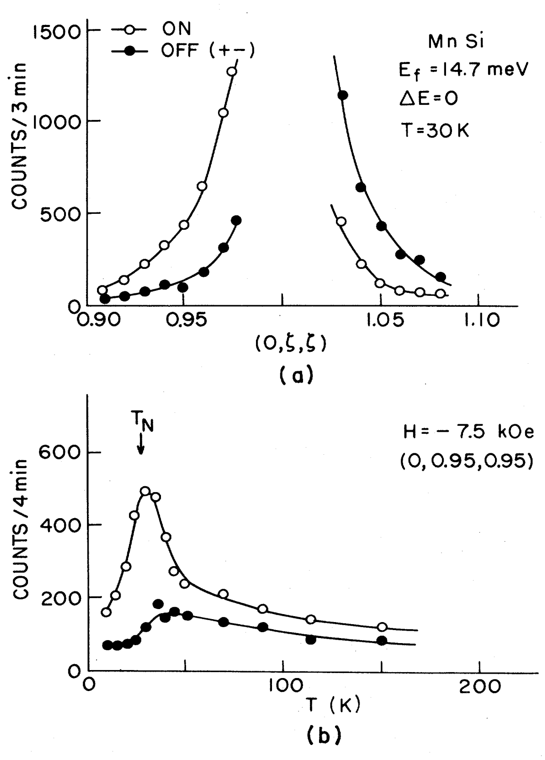

FIG. 4. Polarization dependence of critical scattering from MnSi at T_N and above. Horizontal field is applied along [011]. Note that a significant on−off difference remains at 150 K, 5 times T_N.

III. DIFFUSE SCATTERING IN THE FERROMAGNETIC PHASE AND PARAMAGNETIC PHASE

The major portion of our measurements were carried out under a horizontal magnetic field of ± 7.5 kOe when, as shown in Fig. 1(a), MnSi is a ferromagnet. The experiments were performed with the experimental arrangement I of Fig. 1 with a pyrolytic graphite monochromator and Heusler analyzer which scattered neutrons at energies of 14, 31, or 60 meV. A shown in Fig. 2(b), a normal ferromagnet, such as a Heusler crystal, gives a spin-flip scattering $I^{+-}(\omega)$ which is equal to $I^{-+}(-\omega)$ provided that the frequency transfer ω is much less than $kT/\hbar$. In particular, for $\omega=0$, the two cross sections are equal.

Figure 4 shows the scattering observed from MnSi with the spectrometer set for zero energy transfer $\Delta E=\omega=0$, and clearly there are large differences between the scattering observed with the flipper off $(+-)$ and that with the flipper on $(-+)$, and the differences occur for a range of wave vectors and for temperatures up to at least 150 K. Figure 4(a) shows that the difference of the on and off counts reverses sign on passing through the (110) Bragg reflection, and that the most intense scattering is observed for the same polarization as that found for the Bragg scattering in Fig. 3.

The asymmetry in the scattering for a wave-vector transfer of $\vec{Q}=(0,0.95,0.95)$ is almost independent of field for $T>T_c$, as shown in Fig. 5, except that it changes sign when the field reverses because the direction of rotation of the spin between the applied field at the specimen and at the analyzer is then in the opposite sense.

So far we have described the eleastic neutron scattering. As mentioned in the Introduction, our current study originated from a study of the forbidden magnon scattering. The standard inelastic scattering from a typical ferromagnet should show characteristic patterns as demonstrated in Fig. 2(b). Forbidden magnons are those that appear in the "wrong" channels near and above T_c. We have so far demonstrated that a one-handed sprial ferromagnet exhib-

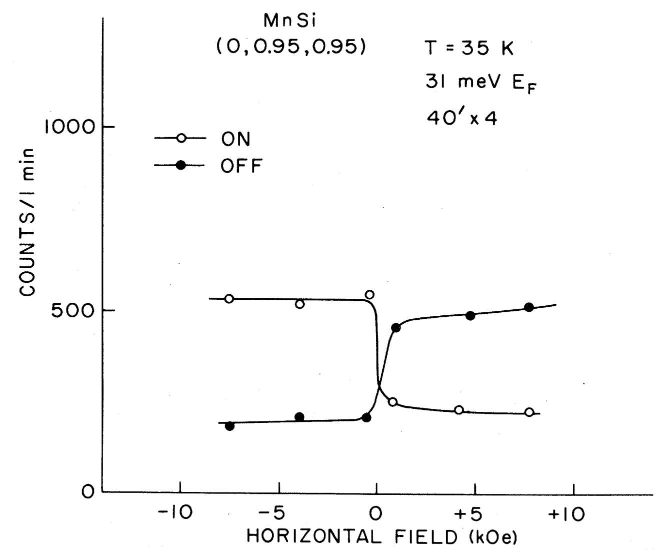

FIG. 5. Field dependence of on and off counts at 35 K.

its a pronounced polarization dependence, which superposes on the pattern in Fig. 2(b). In the limit of very high magnetic field, MnSi should show normal ferromagnetic character. At low temperatures the neutron energy-gain cross section ($-+$ in our notation) is suppressed by the population factor and we expect to see only ($+-$) or off-channel scattering.

The result of a typical inelastic scan is shown in Fig. 6. With a negative applied field, the on ($-+$) cross section is more intense than the off ($+-$) cross section for $\vec{Q}=(0,0.96,0.96)$, and the peak occurs at a slightly smaller energy transfer. The temperature dependence of the cross sections are shown in Fig. 7. The results at 30 K are different from those shown in Fig. 6 because in Fig. 7 the applied field was reversed in direction and because the data was taken under somewhat different conditions. At 60 K, the two peaks have nearly the same energies but their intensities are still quite different.

The detailed balance condition for the spin-flip cross section $I^{+-}(Q,\omega)$ is given by[14]

$$I^{+-}(\vec{Q},\omega)=I^{-+}(-Q,-\omega)\exp(-\beta\hbar\omega) \ . \tag{1}$$

In systems with a center of symmetry $\vec{Q}\to-\vec{Q}$ and $I^{+-}(Q,0)=I^{-+}(Q,0)$, but in systems lacking a center of symmetry, such as MnSi, there is no such simple relationship. The intensity $I^{+-}(Q,\omega)$ is proportional to a spin-spin correlation function $J^{+-}(Q,\omega)$ multiplied by the form factor squared so that

$$I^{+-}(Q,\omega)=|f(Q)|^2 J^{+-}(Q,\omega) \ .$$

The use of translational invariance of the correlation function gives

$$I^{+-}(\vec{\xi}+\vec{q},\omega)=I^{-+}(\vec{\xi}-\vec{q},-\omega)\times\exp(-\beta\hbar\omega)\left|\frac{|f(\vec{\xi}+\vec{q})|^2}{|f(\vec{\xi}-\vec{q})|^2}\right| \ . \tag{2}$$

This result is satisfied by the results shown in Fig. 5.

The correlation functions are related to the static susceptibility using a Kramer-Kronig relationship as

$$\chi^{+-}(q)=\frac{1}{\pi}\int_{-\infty}^{\infty}\frac{1}{\omega}[1-\exp(-\beta\hbar\omega)]J^{+-}(q,\omega)d\omega \ . \tag{3}$$

Further analysis can only be performed on the basis of a model of MnSi. Bak and Jensen[5] have expanded the free energy in powers of the spin density and from this we can obtain the susceptibility. Bak and Jensen show that the free energy is given by

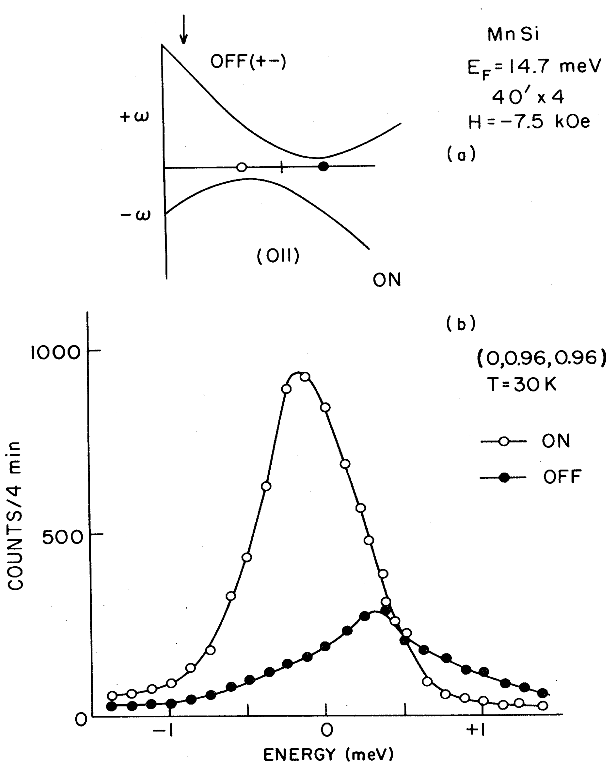

FIG. 6. Inelastic scattering from MnSi at 30 K and at −7.5 kOe. Inset shows schematic model of magnon dispersion curves at the low-temperature limit.

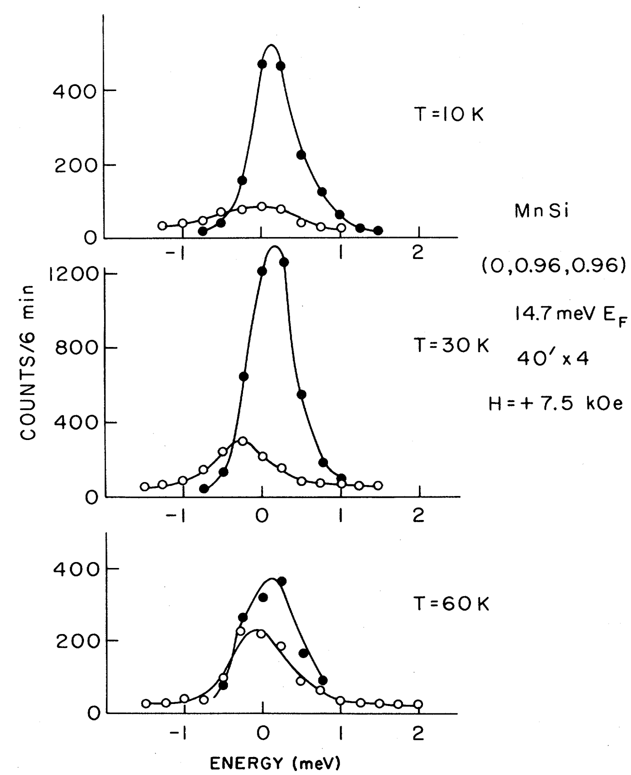

FIG. 7. Inelastic scattering at +7.5 kOe at three different temperatures.

$$F(\vec{r})=\tfrac{1}{2}A(S_x^2+S_y^2+S_z^2)+b\vec{s}(\vec{\nabla}\times\vec{S})+\tfrac{1}{2}B_1[(\vec{\nabla}S_x)^2+(\vec{\nabla}S_y)^2+(\vec{\nabla}S_z)^2]+\tfrac{1}{2}B_2\left[\left[\frac{\partial S_x}{\partial x}\right]^2+\left[\frac{\partial S_y}{\partial y}\right]^2+\left[\frac{\partial S_z}{\partial z}\right]^2\right]$$
$$+C(S_x^2+S_y^2+S_z^2)^2+D(S_x^4+S_y^4+S_z^4)\,, \tag{4}$$

where all of these terms are allowed in most crystals except for the second Dzyaloshinski term which is only allowed in systems lacking a center of symmetry. The use of this free-energy density to calculate the contribution to the free-energy density of an $S^+(q)$ fluctuation along the (110) axis with a magnetic field along that axis gives

$$F(q)=[\tfrac{1}{2}A'-bq+(\tfrac{1}{2}B+\tfrac{1}{4}B_2)q^2]S^+(q)S^-(-q)+[\tfrac{1}{2}A'+bq+(\tfrac{1}{2}B+\tfrac{1}{4}B_2)q^2]S^-(q)S^+(-q)\,, \tag{5}$$

where $A'=A+2c\langle S\rangle^2$ and $A\approx K(T-T_c)$.

Since the coefficients of $S^+(q)S^-(-q)$ gives the inverse susceptibility $[\chi^{+-}(q)]^{-1}$ and that of $S^-(q)S^+(q)$ gives the inverse susceptibility $[\chi^{-+}(q)]^{-1}$, we can now calculate the integral of the scattering cross sections. At high temperatures A is large and there is little difference between $\chi^{+-}(q)$ and $\chi^{-+}(q)$. Upon cooling A becomes smaller and $\chi^{+-}(q)$ has a maximum at the wave vector q, corresponding to the spiral structure, and is very asymmetric with respect to q and $-q$.

If we identify a frequency squared $\omega^2(q)$ with the inverse of the susceptibility the dispersion relations for these frequencies are schematically shown in Fig. 6(a). The asymmetry in the intensity arises from the asymmetry in the $(+-)$ and $(-+)$ dispersion relations. This analysis if broadly consistent with the experimental results but a full theory requires a knowledge of the spin dynamics over a wide range of temperature, which is more complex.

In conclusion we have shown that the lack of symmetry in MnSi gives rise not only to a one handedness in the spiral structure but also to a chirality in the magnetic fluctuations of the ferromagnetic phase over a wide range of wave vectors and temperatures. This chirality can be studied in detail using polarized neutron scattering techniques.

ACKNOWLEDGMENTS

We would like to thank Y. Endoh and O. Steinsvoll for many informative discussions. Work performed at Brookhaven National Laboratory was supported by the Division of Materials Sciences, U. S. Department of Energy, under Contract No. DE-AC02-76CH00016 and at Northwestern University by the National Science Foundation under Grant No. DMR-79-06371.

---

*Permanent address: Department of Physics, University of Edinburgh, Edinburgh EH9 3JZ, Scotland.

[1]Y. Ishikawa, K. Tajima, P. Bloch, and M. Roth, Solid State Commun. 19, 525 (1976).

[2]Y. Ishikawa, G. Shirane, J. Tarvin, and M. Kohgi, Phys. Rev. B 16, 4956 (1977).

[3]Y. Ishikawa, Y. Noda, C. Fincher, and G. Shirane, Phys. Rev. B 25, 254 (1982).

[4]K. R. A. Ziebeck, P. J. Brown, J. G. Booth, and J. A. C. Bland, J. Phys. F 11, L127 (1981); K. R. A. Ziebeck, H. Capellmann, P. J. Brown, and J. G. Booth, Z. Phys. B 48, 241 (1982).

[5]P. Bak and M. H. Jensen, J. Phys. C 13, L881 (1980).

[6]O. Nakamishi, A. Yanase, A. Hasegawa, and M. Kataoka, Solid State Commun. 35, 995 (1980).

[7]A. Yoshimori, J. Phys. Soc. Jpn. 14, 807 (1959).

[8]M. Ishida, Y. Endoh, Y. Ishikawa, S. Mitsuda, and M. Tanaka (private communication).

[9]J. B. Sokoloff, C. H. Perry, R. D. Lowde, F. Soffge, V. Wagner, and M. C. K. Wiltshire, J. Appl. Phys. 50, 1961 (1979).

[10]J. B. Sokoloff, B. Pagonis, C. H. Perry, C. F. Majkrzak, G. Shirane, and Y. Ishikawa (unpublished).

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

[12]G. Shirane, J. D. Axe, C. F. Majkrzak, and T. Mizoguchi, Phys. Rev. B 26, 2575 (1982).

[13]M. Blume, Phys. Rev. 130, 1670 (1963).

[14]D. McK Paul, R. A. Cowley, W. G. Stirling, N. Cowlam, and H. A. Davis, J. Phys. F 12, 2687 (1982).

[15]M. Kataoka, J. Magn. Magn. Mater. 31-34, 341 (1983).