PHYSICAL REVIEW B VOLUME 18, NUMBER 9 1 NOVEMBER 1978

Field dependence of spin dynamics in the itinerant ferromagnet MnSi

J. A. Tarvin,* G. Shirane, and Y. Endoh†
Brookhaven National Laboratory, Upton, New York 11973

Y. Ishikawa
Physics Department, Tohoku University, Sendai, Japan

(Received 17 May 1978)

Inelastic neutron scattering has been used to study the field dependence of low-energy magnetic excitations of the weak itinerant ferromagnet MnSi. At 5 K, the gap in the spin-wave dispersion relation increases linearly with applied magnetic field, with a slope that is consistent with a *g* factor of 2, and the spin-wave stiffness is independent of field. At 45 K, which is 15 K above the zero-field T_c, a 70-kOe field restores spin-wave behavior with a gap that is nearly equal to that at 5 K and with a stiffness about half as great as that observed at low temperature. The Stoner boundary seems to be shifted by the field, especially above T_c.

I. INTRODUCTION

In a previous paper[1] (hereafter referred to as I), we presented neutron-scattering measurements which demonstrate that the magnetic excitations in MnSi have the properties expected in a weak itinerant ferromagnet. This discovery is fortunate, for the characteristic energy of magnetic excitations in MnSi is well within the range which can be studied by inelastic neutron scattering; and, for the first time, it is possible to study the spin dynamics of an itinerant ferromagnet thoroughly.

As shown schematically in Fig. 1, there are two distinct contributions to the inelastic magnetic scattering in MnSi. Because single-particle excitations are forbidden in region I, well-defined spin waves which follow a quadratic dispersion relation are permitted there. These collective excitations renormalize and broaden with increasing temperature. The intensity of scattered neutrons, integrated over energy at a given momentum, decreases rapidly as the Stoner boundary is crossed; and, above the Stoner boundary in region II, the excitations are very broad and relatively independent of temperature, as expected for interband transitions[2] in the Stoner continuum. In the limit of a noninteracting electron gas, these would be single-particle excitations, but the existence of a weak interaction gives some collective character to excitations in the Stoner continuum. The random-phase approximation, applied to a weakly interacting electron-gas model, provides a good qualitative explanation of the observed features of the magnetic excitations in MnSi.

A third region, lying between I and II in Fig. 1, has been observed in Fe (Ref. 3) and Ni.[4] In this region, the excitations are very similar to spin waves; they broaden with increasing temperature and do not show the dramatic decrease in intensity observed in the vicinity of the Stoner boundary. However, the energy of these excitations remains finite at and above T_c. Their existence has been explained on the basis of a short-range magnetic order that persists above T_c.[5] There is no direct evidence of these intermediate excitations, which should not be confused with excitations in the Stoner continuum, in MnSi.

In this paper, we extend I by studying the effect of a magnetic field on the excitations at and below

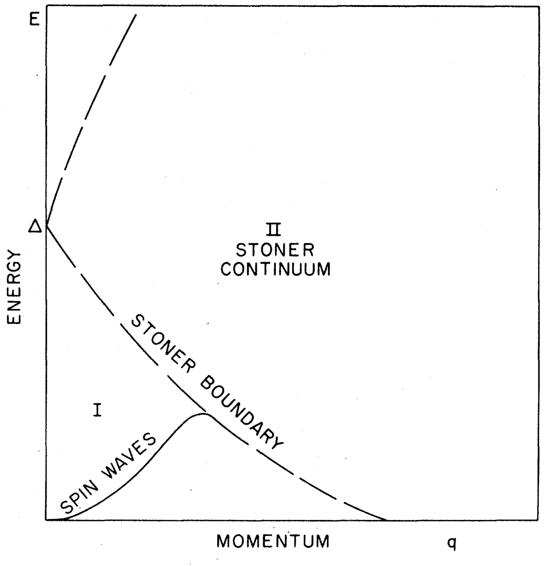

FIG. 1. Schematic diagram of the magnetic excitations in MnSi. Because single-particle excitations are forbidden in region I, there are spin waves which follow the dispersion relation described by the solid line. The excitations in the Stoner continuum are very broad because they have the character of single particles. The energy Δ at which the Stoner boundary intersects the energy axis is the splitting between electron bands that have opposite spin.

the Stoner boundary. In Sec. II we mention some relevant points concerning the crystal structure of MnSi and the sample used in this experiment. We also discuss some details of the experiment. In Sec. III we present the results of our experiments performed at 5 and 45 K.

II. EXPERIMENTAL DETAILS

Manganese silicide has the cubic B-20 crystal structure with $a = 4.553$ Å and eight atoms per unit cell. In the absence of a field, it orders magnetically with a helical spin structure[6] below $T_c = 30$ K. The screw axis is the [111] crystallographic direction and the period is 180 Å. In an applied field of 6 kOe, the moments align in an induced ferromagnetic state.[7-9] It is in this phase that ferromagnetic spin waves are observed. The excitations in the Stoner continuum are observed even in zero field.

The sample used in this experiment was grown from the melt by the Czochralski method by Miura at the Institute for Iron, Steel, and other Metals at Tohoku University. It is a cylinder 20 mm in diameter and 40 mm long, with $[01\bar{1}]$ nearly parallel to the cylindrical axis. Parts of this crystal, which was used in the experiment detailed in I, have been used for measurements of ultrasonic attenuation,[9] electron spin resonance[10] (ESR), and small-angle neutron diffraction.[6]

The experiment was carried out on a triple-axis neutron spectrometer at the Brookhaven high-flux beam reactor. A pyrolytic graphite (002) monochromator and analyzer were used. Most of the data were taken with the *outgoing* neutron energy (E_f) fixed at 14.8 meV ($14.8E_f$), and with the four horizontal collimators set at 40′-20′-40′-40′ full width at half-maximum (FWHM). Some additional data were taken with the *incoming* energy (E_i) fixed at 14.8 meV ($14.8E_i$), and with collimators set at 40′-40′-40′-40′ or 20′-20′-20′-40′. For $14.8E_i$, a graphite filter was placed before the monochromator to remove higher-energy neutrons from the beam. For $14.8E_f$, a filter was inserted after the analyzer.

The crystal was mounted with $[01\bar{1}]$ vertical in a variable-temperature cryostat inside a vertical-axis superconducting magnet. Most of the data were taken in the vicinity of the 011 reciprocal-lattice point because of the relatively large ratio of the magnetic to the nuclear structure factor at 011. Some data were also taken near 000. The [111] direction was chosen to eliminate the spurious data points occasionally seen in the [100] direction.

III. RESULTS

A. Field dependence at low temperature

In a Heisenberg ferromagnet, the effect of a magnetic field H at low temperature is to increase the gap in the spin-wave dispersion relation.[11] The spin-wave energy then becomes

$$E = \Delta_{sw} + Dq^2, \quad \Delta_{sw} = \Delta_0 + g\mu_B H, \tag{1}$$

where D is the spin-wave stiffness, q is the momentum of the excitation, g is the Landé g factor, and μ_B is the Bohr magneton. Δ_0, which is due to anisotropy fields,[11] is negligible in MnSi. Note that Δ_{sw} is not to be confused with the band split-

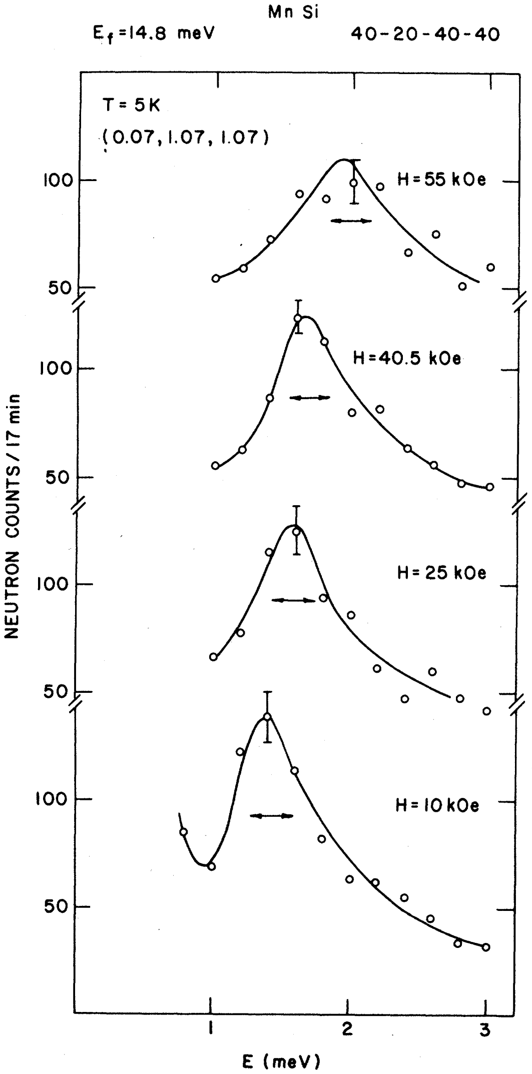

FIG. 2. Field dependence of a spin-wave group propagating along [111]. The lines are smooth curves drawn through the data and the double arrows indicate the FWHM of the spectrometer resolution function as calculated from Refs. 12 and 13.

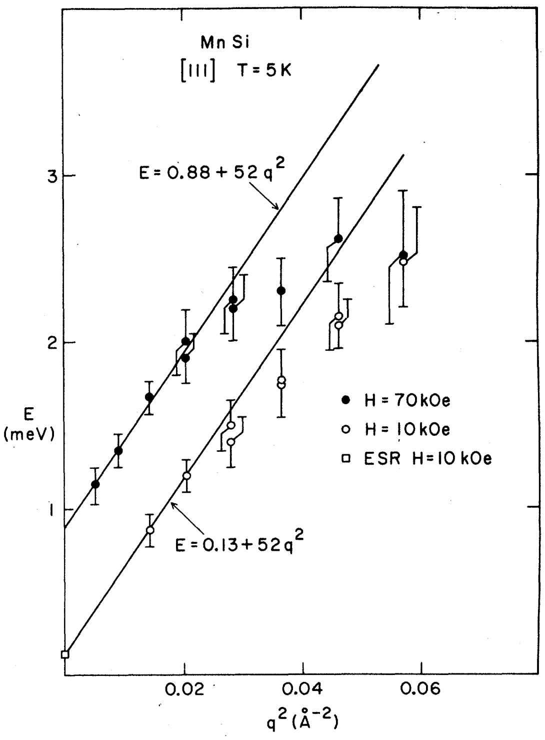

FIG. 3. Dispersion relation for [111] spin waves in low and high fields. Note that the first deviation from a q^2 relation [Eq. (1)] occurs near $q^2 = 0.03$ Å^{-2} in both cases, and that the two dispersion curves meet in the Stoner continuum ($q^2 \gtrsim 0.06$ Å^{-2}).

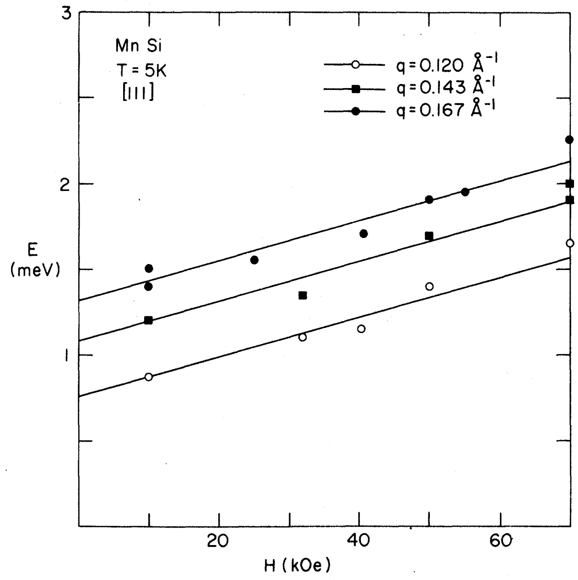

FIG. 4. Field dependence of the excitation energy for three different momentum transfers in the spin-wave region. The slope of the lines is determined by $g = 2.0$.

ting Δ (Fig. 1). Figure 2 shows how an increasing magnetic field affects excitations in the spin-wave region. The double arrows indicate the FWHM of the spectrometer resolution function as calculated from the formulas of Cooper and Nathans[12] and Chesser and Axe.[13] The excitation energy gradually increases with increasing field while the linewidth remains nearly constant. The spin-wave dispersion relation is depicted for fields of 10 and 70 kOe in Fig. 3. The spin-wave stiffness of 52 meV Å^2 is consistent with [100] data presented in I. Along [111], however, the first departure from q^2 dependence [Eq. (1)] occurs at $q^2 \approx 0.03$ Å^{-2} (independent of field), while the departure appears at $q^2 \approx 0.05$ Å^{-2} along [100]. Above $q^2 = 0.03$ Å^{-2}, the energy shift produced by the field decreases until there is no visible shift at $q^2 = 0.06$ Å^{-2} in agreement with the observation that the excitations in the Stoner continuum are not modified appreciably by the external field.

An ESR measurement[10] has determined that g is 2.0 MnSi. In that case, $g\mu_B \times 60$ kOe is 0.70 meV, a number which compares well with the 0.75-meV change in gap energy from 10 to 70 kOe in Fig. 3. In Fig. 4, the spin-wave energy is plotted as a function of field for three different wave vectors. In each case, the data are consistent with the solid lines, which have a slope corresponding to $g = 2.0$.

In order to determine the position of the Stoner boundary and to show clearly the effect of the field in its vicinity, many scans were made for momentum transfer in the [111] direction about the 011 reciprocal-lattice point with a field of 70 kOe. A contour plot made from those scans is shown in Fig. 5. A rapid broadening, which is indicative of the Stoner boundary, occurs at $E_{70} = 2.6$ meV and $\zeta_{70} = 0.09$ ($q_{70} = 0.22$ Å^{-1}, $q_{70}^2 = 0.05$ Å^{-2}). The broadening is more distinct at high field in the [111] direction because the peak intensity in the spin-wave region is shifted to higher energy and there is less overlap of inelastic scattering (signal) with elastic incoherent scattering (background).

Figure 11(b) of I shows that the boundary is at $E_{10} = 2.4$ meV and $q_{10} = 0.25$ Å^{-1} for a 10-kOe field. E_{10} and E_{70} lie within errors of each other, but the momenta at the boundary are better defined and the difference between q_{10} and q_{70} is significant. This result suggests that the shift of the Stoner boundary by the application of a high field, if it exists, is much smaller than that of the spin waves.

In the case of a weak itinerant ferromagnet, where the simple band theory works as shown in I, both the Stoner boundary and the spin-wave stiffness are proportional to the magnetization. The increase in magnetization[14] induced by the high field at low temperature in MnSi is large when

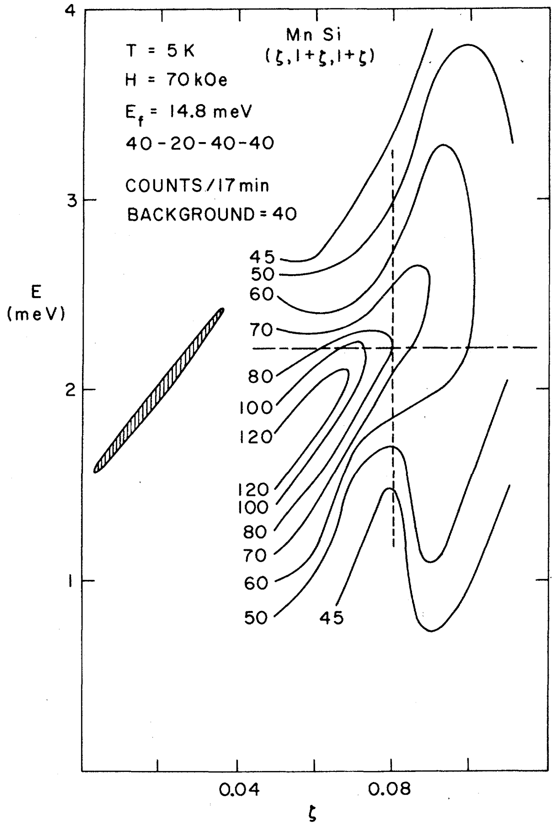

FIG. 5. Contour plot of the intensity of inelastic scattering in a high field as a function of energy and momentum. The rapid broadening near the Stoner boundary, visible only for [100] in a low field, is distinct here for [111]. The peak in a constant-q scan (dashed line) may not coincide with the peak in a constant-E scan (dot-dashed line). See text. Momentum is expressed in the reduced unit ζ, which is defined so that $\vec{q} = (\zeta, \zeta, \zeta)(2\pi/a)$.

compared to the increase that would be expected in a Heisenberg system, but it is still a relatively small fraction of the total magnetization. Consequently, the fact that neither the spin-wave stiffness nor the Stoner boundary is sensitive to the magnetic field at low temperature is consistent with the band model.

In principle, we can determine the Stoner boundary over a wide range in q and ω by shifting the spin-wave dispersion continuously with an external field. The accuracy of our present experiments is, however, not sufficient to provide useful information.

Figure 5 also shows that a contour plot supplies much more information than a compilation of peak positions and linewidths. The path of a "constant-q" scan for $\zeta = 0.08$ is given by the dashed line. The peak is at $E = 2.2$ meV. The dot-dashed line shows the path of a "constant-E" scan for $E = 2.2$ meV. Its peak occurs not at $\zeta = 0.08$ but at $\zeta = 0.07$.

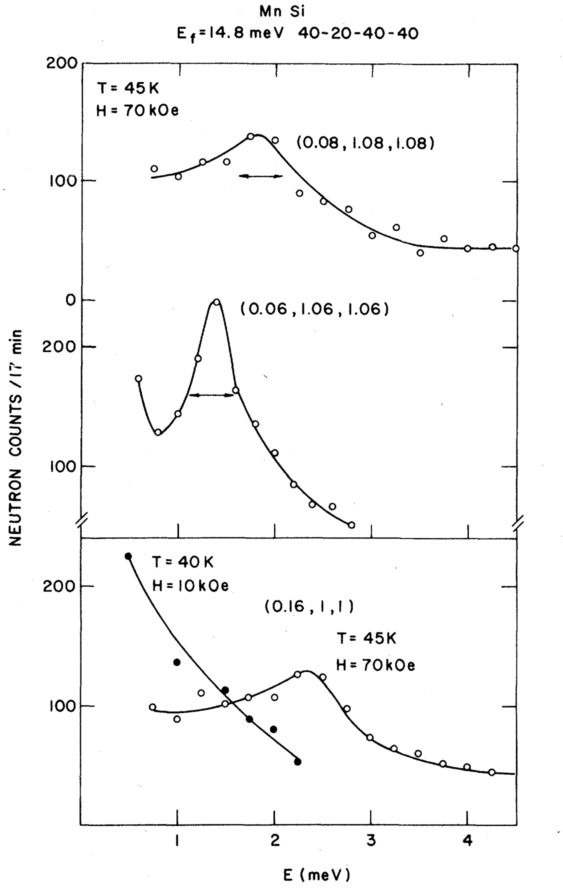

FIG. 6. Spin-wave groups above T_c restored by a 70 kOe field. Observe that in a field of 10 kOe the spin-wave scattering has merged with the critical scattering (filled circles).

This disagreement between constant-q and constant-E scans can occur whenever the measured cross section has a significant linewidth. Even computer fitting cannot resolve the ambiguity unless the exact form of the dynamic structure factor $S(q, \omega)$ is known.

B. Field dependence above T_c

As described in I, spin waves are not visible in MnSi at temperatures above 30 K in a field of 10 kOe. In 70 kOe, however, spin waves are quite distinct even at 45 K, as can be seen from Fig. 6, where [111] scans are shown for high field and [100] scans for both low and high fields. Notice that the high field *quenches the critical scattering and restores spin waves along* [*100*]. The dispersion relation at 45 K and 70 kOe is plotted in Fig. 7. The gap is only slightly smaller than it is at 5 K, and the region over which Eq. (1) holds is larger in Fig. 7 than it is in Fig. 3.

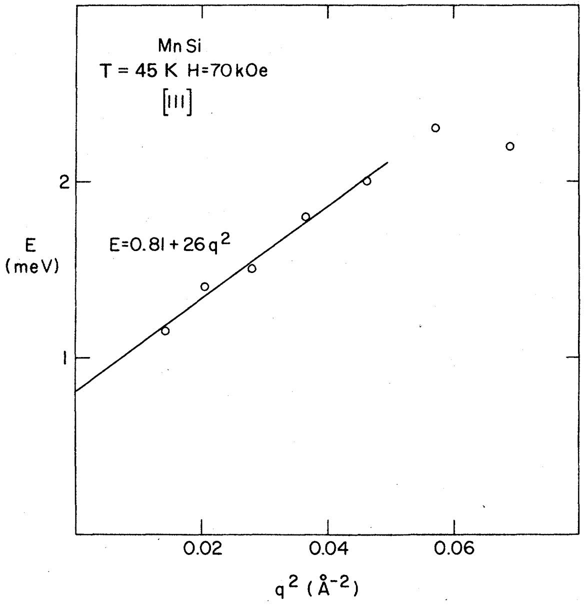

FIG. 7. Spin-wave dispersion relation in 70 kOe above T_c.

A contour plot of this data does not show an abrupt broadening, and gives no clear indication of the position of the Stoner boundary. One can see from Fig. 6 that the boundary definitely lies above $\zeta = 0.06$ and may lie above 0.08 along [111]. It is probably below $\zeta = 0.10$, since the excitation is very broad there and is very similar to the low-field, low-temperature excitation. So the momentum at the boundary is about the same at 45 as at 5 K. Since the energy is about 2.2 as opposed to 2.6 meV, we can conclude that the energy at the Stoner boundary E_{SB} ($T = 45$ K, $H = 70$) is about 80% of E_{SB} ($T = 5$ K, $H = 70$). The result is again consistent with the conjecture that the Stoner boundary is proportional to the magnetization, because the magnetization M ($T = 45$ K, $H = 70$ kOe) is 70% of M (5 K, 70 kOe).[14]

Note that the spin-wave stiffness D ($T = 45$ K, $H = 70$ kOe) is only half of D (5 K, 70 kOe). If the spin waves are renormalized by magnon-magnon interactions, as in other systems,[11,15] D should decrease more slowly than M with increasing temperature. The opposite result here suggests that other renormalization mechanisms—possibly magnon-electron interactions—are important. However, a detailed comparison cannot be made with other systems, since no detailed study has been made previously on spin waves above T_c in such a high field.

C. Discussion

Because there is no theory with which we can compare our data directly, our remarks here will be brief. The actual field dependence of the magnetic excitations in MnSi matches the qualitative expectations. At 5 K, the gap in the spin-wave spectrum is proportional to the field, the g factor is 2, and there is no change in the spin-wave stiffness. We are not able to determine definitely whether the position of the Stoner boundary is changed by the field, but the results are not inconsistent with the simple band picture that the Stoner boundary varies with temperature (Figs. 12 and 13 in I) and with field in proportion to the magnetization. Using the band model, we can understand the fact that the spin waves are restored by the field, because at 45 K in a field of 70 kOe, the Stoner boundary is elevated almost to its value at low temperature and the spin-wave region I in Fig. 1 reappears even above T_c. Since the direct temperature effect on $\chi(Q, \omega)$ is rather small, the spin waves can reappear in region I.

Note that we cannot exclude the possibility that the Stoner boundary is insensitive to temperature and field as observed in Ni (Ref. 4) and Fe.[3] However, in this case we need to assume that the magnetic interactions in MnSi are quite anisotropic, and that short-range order with long correlations remains even above T_c in order to explain the strong field dependence at 45 K. This is the situation realized in low-dimensional material,[16] but is difficult to expect for cubic MnSi.

ACKNOWLEDGMENTS

We wish to thank R. H. Swendsen, J. N. Fields, T. Moriya, and T. Kasuya for helpful discussions. Research for this paper was supported by the Division of Basic Energy Sciences, Department of Energy, under Contract No. EY-76-C-02-0016.

---

*Present address: KMS Fusion, Inc., Ann Arbor, Mich. 48106.

†Permanent address: Physics Dept., Tohoku University, Sendai, Japan.

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

[2] W. Marshall and S. W. Lovesey, *Theory of Thermal Neutron Scattering* (Oxford University, N. Y., 1971), Chap. 9.

[3] J. W. Lynn, Phys. Rev. B 11, 2624 (1975).

[4] H. A. Mook, J. W. Lynn, and R. M. Nicklow, Phys. Rev. Lett. 30, 556 (1973).

[5] V. Korenman, J. L. Murray, and R. E. Prange, Phys. Rev. B 16, 4048 (1977).

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

[7] H. J. Williams, J. H. Wernick, R. C. Sherwood, and G. K. Wertheim, J. Appl. Phys. 37, 1256 (1966).

[8] D. Shinoda and S. Asanabe, J. Phys. Soc. Jpn. 21, 555 (1966).

[9] S. Kusaka, Y. Yamamoto, T. Komatsubara, and Y. Ishikawa, Solid State Commun. 20, 925 (1976).

[10] M. Date, K. Okuda, and K. Kadowaki, J. Phys. Soc. Jpn. 42, 1555 (1977).

[11] F. Keffer, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1966), Vol. 18, P. 2.

[12] M. J. Cooper and R. Nathans, Acta Crystallogr. 23, 357 (1967).

[13] N. J. Chesser and J. D. Axe, Acta Crystallogr. A 29, 160 (1973).

[14] D. Bloch, J. Voiron, V. Jaccarino, and J. H. Wernick, Phys. Lett. A 51, 259 (1975).

[15] J. A. Tarvin, R. J. Birgeneau, G. Shirane, and H. S. Chen, Phys. Rev. B 17, 241 (1978).

[16] J. Als-Nielsen, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1976), Vol. 5A, p. 147.