

TABLE III. Symmetry permitted configurations in some of the Shubnikov groups  $Pmc2_1$ . The primes denote antisymmetry operations. The unit cell vectors refer to the tetragonal unit cell. The orientation of the orthorhombic cell is defined in the text.

	$a$	$b$	$c$
$Pmc2_1$	$A$	$G$	$C$
$Pm'c2_1'$	$F$	$C$	$G$
$Pmc'2_1'$	$G$	$A$	$F$
$Pm'c'2_1$	$C$	$F$	$A$

subgroups of  $P4_2/mbc$ , namely,  $Pbam$ ,  $Pba2$ ,  $Pmc2_1$ , and  $P2_12_12$ , the point symmetry of the  $Fe^{2+}$  sites confines the spin directions to along an axis or within a plane in all cases except  $Pmc2_1$ . The symmetry permitted configurations in the set of Shubnikov groups<sup>7</sup>  $Pmc2_1$  for those which do not involve an enlarged unit cell are given in Table III. The orthorhombic cell has the orientation  $a'=c$ ,  $b'=a$ ,  $c'=b$ , where  $a$ ,  $b$ , and  $c$  are the tetragonal unit cell vectors, the latter having been used in Table III. The origin is shifted by  $(0, \frac{1}{4}, 0)$ , and the  $Fe^{2+}$  ions occupy the general positions  $x, y, z$ . For  $x=\frac{1}{4}$ ,  $y=\frac{1}{4}$ , and  $z=\frac{1}{2}$ , their relative positions are un-

<sup>7</sup> N. V. Belov, N. N. Neronova, and T. S. Smirnova, *Kristallografiya* **2**, 315 (1957) [English transl.: *Soviet Phys.—Cryst.* **2**, 311 (1957)].

changed. From this viewpoint, the model  $A_zG_yC_z$  is favored (Shubnikov group  $Pmc2_1$ , No. 26-66<sup>7</sup>) in which case the components  $G_y$  and  $C_z$  have moments of  $(0.9 \pm 0.2)\mu_B$  and  $(0.7 \pm 0.2)\mu_B$ , respectively.

The crystal structure at 4.2°K should reflect the orthorhombic symmetry, and a diffractometer trace at 4.2°K did in fact reveal that the  $(hkl)$  peaks in general and  $(h00)$  peaks in particular were visibly broadened, while  $(hh0)$  and  $(hhl)$  peaks were not. The resolution was not sufficient for the distortion to be measured accurately, but at a rough estimate there is a difference of 0.2% between the original tetragonal  $a$  axes. However, this effect was still present well above 77°K, and hence the distortion is not connected with the magnetic transition.

It is interesting to note that in orthorhombic  $\beta$ - $CoSO_4$  the combination of antiferromagnetic modes  $A_xG_yC_z$  has been definitely established at 4.2°K.<sup>8,9</sup> The crystal structures of this compound and  $FeSb_2O_4$  can be considered analogous to the extent that the  $3d$  ions occupy similar sites within chains of oxygen octahedra along the  $c$  axes.

<sup>8</sup> P. J. Brown and B. C. Frazer, *Phys. Rev.* **129**, 1145 (1963).

<sup>9</sup> R. Ballestracci, E. F. Bertaut, J. Coing-Boyat, A. Delapalme, W. James, R. Lemaire, R. Pauthenet, and G. Roult, *J. Appl. Phys.* **34**, 1333 (1963).

## Local Antiferromagnetic Order in Single-Crystal MnO above the Néel Temperature\*

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This investigation of the spin arrangement in a single crystal of MnO above the Néel temperature ( $T_N \simeq 122^\circ\text{K}$ ) has shown that the magnetic neutron scattering in the vicinity of the (111) magnetic peak position consists of diffuse but distinct satellites. The presence of these satellites, which are lined up approximately along a  $\langle 111 \rangle$  axis of the single crystal, indicates that small regions containing antiphase boundaries and strong antiferromagnetic order exist well above  $T_N$ . Applying a model developed for domains in ordered alloys, the average domain size was estimated to be  $46 \pm 5 \text{ \AA}$  at 133°K. The general features of the scattering indicate that the number of coherent domains in a given region is small, and lead to the preliminary conclusion that the local order in MnO above  $T_N$  is inhomogeneous.

### I. INTRODUCTION

A RECENT study of diffuse magnetic neutron scattering in powder samples of MnO<sup>1</sup> showed that there is a local coupling above the Néel temperature ( $T_N \simeq 122^\circ\text{K}$ ) with spins tending to remain parallel to a  $\{111\}$  plane, and with neighboring planes arranged in an antiferromagnetic fashion. It was also apparent from that study that the directional preference for a  $\{111\}$  layering of the spins diminished as the tem-

perature increased, so that at room temperature the fit of the diffuse data to a (111) model was less satisfactory than at temperatures just above  $T_N$ . The local correlations were estimated to extend over regions of 20–50 Å.

The current work on a single crystal of MnO presents a more detailed examination of the neutron spin scattering from this material, and this provides a much better insight into the nature of the local spin order above the critical point. The powder method yields a spherical average of diffuse scattering for each diffraction vector; the single-crystal method, on the other

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<sup>1</sup> I. A. Plech and B. L. Averbach, *Physics* **1**, 31 (1965).

hand, enables us to observe the distribution of scattering in reciprocal space. This study examines the scattering in the (110) plane of reciprocal space in the vicinity of the (111) position, as defined by the face centered cubic magnetic cell with lattice constant,  $a_0 = 8.85 \text{ \AA}$ . Below the critical temperature there is a strong coherent magnetic peak at (111) which is characteristic of the long-range order. The distribution of the diffuse magnetic scattering above  $T_N$  is characteristic of the local order above the critical temperature, and we have focused our attention on the form of this diffuse intensity.

It appears that the long-range spin order in MnO is never perfect, even at liquid-helium temperatures and, since both the long-range and short-range order are developed with spins parallel to  $\{111\}$  planes, it has been postulated that large antiphase domains are frozen into the ordered structure.<sup>2</sup> Pershan<sup>3</sup> has reported the absence of antiferromagnetic domain walls in ordered  $\text{MnF}_2$  from observations of the  $F^{19}$  nuclear resonance under various combinations of applied electric and magnetic fields. Pershan also pointed out that antiferromagnetic domains may exist just below the Néel point, but these domains are thermodynamically unstable and should grow out at lower temperatures unless pinned by lattice defects. Similar reasoning may be applied to MnO and, by extension, to MnO above the critical temperature. From the work reported here, it appears that domains with short correlation ranges may be stable above  $T_N$ , even though the crystal has undergone cooperative spin disordering. We shall also devote some attention to the analogous behavior in alloy systems undergoing order-disorder transitions.

## II. EXPERIMENTAL RESULTS

The thermal neutrons used in this experiment were monochromated at  $\lambda = 1.20 \text{ \AA}$  by a (111) reflection from a lead single crystal. The  $\lambda/2$  contribution was approximately 5% of the main nuclear peak, and this was eliminated by making measurements with a Pu absorber.

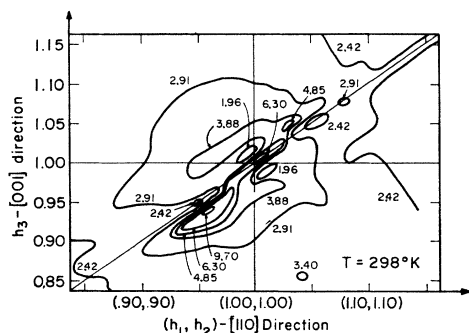


FIG. 1. Magnetic scattering in the (110) reciprocal lattice plane near the (111) spot at 298°K. Intensities are in barn/sr.

<sup>2</sup> I. A. Blech and B. L. Averbach, Phys. Rev. **142**, 287 (1966).

<sup>3</sup> P. S. Pershan, Phys. Rev. Letters **7**, 280 (1961).

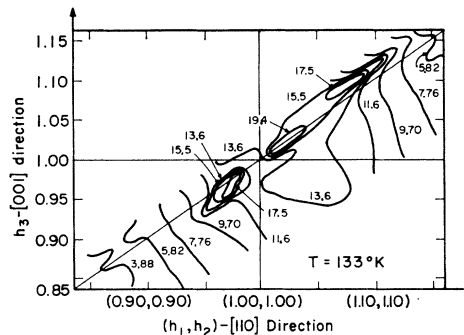


FIG. 2. Magnetic scattering in the (110) reciprocal lattice plane near the (111) spot at 133°K. Intensities are in barn/sr.

The single crystal was mounted on a goniometer within a liquid-nitrogen cryostat. The growth axis of the crystal was a  $\langle 110 \rangle$  direction, and it was thus possible to use a (111) plane parallel to the long axis of the crystal cylinder. The entrance and receiving slits defined a horizontal divergence of  $\pm 15'$  and a vertical divergence of about  $\pm 20'$ .

The resultant diffuse magnetic intensities were corrected for background scattering and for thermal diffuse scattering, using the Debye approximate form for this contribution and an average Debye temperature of 502°K.<sup>4</sup> The nuclear spin incoherent scattering was evaluated, using the cross sections listed by Bacon.<sup>5</sup> An absorption correction for the particular sample shape was determined experimentally, and the multiple scattering correction for this orientation of the crystal was negligible. The intensities were converted into absolute units by comparison with the known incoherent scattering from vanadium.

Inasmuch as the present evaluation of these data are primarily concerned with geometric considerations such as the shape and distribution of diffuse intensity in reciprocal space, the question of inelastic contributions is of secondary importance. We have converted our intensities to absolute units in order to assess accurately the relative magnitude of the fully corrected data, and it is unlikely that the conclusions will be affected by the presence of inelastic magnetic scattering.

The standardized magnetic intensities are shown in Fig. 1 (298°K) and 2 (133°K) as equi-intensity contour maps on a (110) plane in the reciprocal lattice. In these figures the coordinate  $h_3 = 1.00$  refers to a distance of one reciprocal lattice unit,  $1/a_0$ , where  $a_0$  is the magnetic cell lattice parameter (8.85 Å at 298°K). The position (1.00, 1.00, 1.00) thus refers to the (111) magnetic reflection. Each figure clearly shows a diffuse peak close to, but not precisely at, the (111) position. The displacement of this central peak is not an experimental error, since the (111) position is accurately

<sup>4</sup> N. Kuriyama and F. Hosoya, J. Phys. Soc. Japan **17**, 1022 (1962).

<sup>5</sup> G. E. Bacon, *Neutron Diffraction* (Oxford University Press, London, 1962).

defined by the measurement of the (222) nuclear peak, which is not shown. In addition, each diffuse peak is flanked by two satellite peaks approximately along the [111] direction. At 298°K, the lower spot is somewhat more intense than the central peak and separated from it by 0.0915 reciprocal lattice units (rlu). The upper spot is slightly less intense and separated by 0.0549 rlu. The axis through the satellites and the central spot is tilted relative to the [111] by about 5 deg. At 133°K, the satellites are much more clearly defined. The lower spot is separated from the central spot by 0.090 rlu and the upper spot by 0.100 rlu. The two satellites have about the same intensity, but both are slightly less intense than the central peak, with a relative intensity ratio of 0.9. The central spot at 133°K is shifted outward from the (111) by 0.043 rlu, and the satellite axis is tilted by about 2 deg to the [111] direction.

### III. DISCUSSION

The principal characteristics of the diffuse magnetic scattering may be listed as follows; the presence of satellites flanking the main diffuse (111) magnetic peak: the general directionality of the diffuse streaking and its tendency to follow a [111] axis: the displacement of the approximate centroid of the diffuse scattering to an angle larger than that given by the average magnetic cell parameter; and the small tilting of the axis of directional scattering away from the average [111]. In the diffuse scattering from binary alloys which undergo order-disorder transitions, streaking in reciprocal space is associated with a directional disordering, and the appearance of satellites is interpreted by models which involve a quasiperiodic local order in the alloy. The existence of diffuse satellites above the critical ordering temperature has received much attention, and has been observed, for example, in the alloy Cu<sub>3</sub>Au by electron diffraction<sup>6,7</sup> and by x-ray diffraction.<sup>8</sup> These satellites flank the superstructure peak positions, in analogy with the magnetic superstructure positions in MnO, and are consistent with a model of the short-range order involving the formation of a large number of antiphase boundaries in the ordered structure. In this connection, it should be noted that the ordered structure of MnO is analogous to the ordered structure of the alloy CuPt.<sup>9</sup> The spins are parallel within a given (111) layer, but are oppositely directed in alternate (111) layers: in CuPt, (111) planes are alternately occupied by Cu and Pt atoms.

It is convenient to transform the ordered cubic unit cell into a hexagonal cell with a  $c$  axis normal to the (111) planes. This yields a unit cell with  $a_1 = a_2 = \sqrt{2}a_0$ ,

and  $a_3 = c = \sqrt{3}a_0$ . The hexagonal cell contains 24 Mn ions in six close-packed planes: each plane has four Mn ions with parallel spins with alternate spin directions in successive planes. The (111) reflection of the cubic structure becomes the (00.3) hexagonal reflection, and the cubic <111> direction corresponds to the <00.l> axis. The relationship between the magnetic structure factors is given by

MnO (cubic):

$$F_m = 32\phi, \quad h, k, l \text{ all odd, with } h+k, \\ k+l, l+h = 4n \pm 2;$$

$$F_m = 0, \quad \text{otherwise};$$

MnO (hexagonal (00.l) reflections):

$$F_m(00.l) = 24\phi, \quad l = 6n \pm 3; \quad (1)$$

$$F_m(00.l) = 0, \quad \text{otherwise},$$

where  $\phi$  is the magnetic structure amplitude for neutron scattering<sup>5</sup>; the amplitude changes sign as the spin direction reverses. Since the magnetic intensity involves only the component of the spin normal to the diffraction vector, the intensity in the vicinity of a given (00.l) reflection will come mainly from spins which are parallel to that particular plane. Contributions from spins along other {00.l} planes will be negligible, and we may thus treat the problem as a one-dimensional modulation of the local spin order.

We now achieve the local order by a process which involves the disordering of the MnO spin superstructure by slicing up the ordered structure into small regions until we have achieved something like the locally ordered configuration. We do not concern ourselves here with the correlation functions, but rather with the intensity distribution. We shall describe the disordered MnO structure as one in which the ordered structure along [00.l] is interrupted by spin antiphase boundaries. A typical faulted spin layer sequence might then be,  $\uparrow\uparrow\uparrow\uparrow\uparrow\uparrow\downarrow\downarrow\downarrow\downarrow$ , with the indicated antiphase boundary. This sequence creates two adjacent layers of like spin and would seem to produce an excess of ferromagnetically coupled neighbors. However, in order to preserve the net paramagnetism, we must also create two similar layers which are parallel in the opposite sense.

The effect of introducing these mistakes, or antiphase boundaries, in a perfectly periodic fashion has been worked out for several types of ordering alloys.<sup>6,10</sup> Using this notation, we may write the scattered amplitude distribution  $G$  in reciprocal space arising from one-dimensional antiphase boundaries normal to [00.l] as

$$G = \frac{\sin\pi M_1 A_1 \sin\pi M_2 A_2}{\sin\pi A_1 \sin\pi A_2} \\ \times \frac{\sin\pi M_3 A_3 \sin\pi N_3 (M_3 A_3 - \phi)}{\sin\pi A_3 \sin\pi (M_3 A_3 - \phi)}. \quad (2)$$

<sup>6</sup> H. Raether, Z. Angew. Phys. 4, 53 (1952).

<sup>7</sup> M. S. Marcinkowski and L. Zwell, Acta Met. 11, 373 (1963).

<sup>8</sup> S. C. Moss, in *Local Atomic Arrangements Studied by X-ray Diffraction*, edited by J. B. Cohen and J. E. Hilliard (Gordon and Breach Science Publishers, Inc., New York, to be published).

<sup>9</sup> C. B. Walker, J. Appl. Phys. 23, 118 (1952).

<sup>10</sup> H. Sato and R. S. Toth, Phys. Rev. 127, 469 (1962).

$M_1$  and  $M_2$  are the number of hexagonal unit cells along the  $a_1$  and  $a_2$  axis.  $M_3$  is the number of cells along  $[00.l]$  within a single domain and  $N_3$  is the number of such domains that maintain a coherent periodicity.  $N_3$  is thus the number of domains of  $M_3$  cells that follow each other in a regular sequence. There may be many of these ordered sequences in a given crystal, and the diffraction effects must then be summed independently over all of these. The interference function  $G$  has simultaneous maxima under the following conditions, with the  $A_i$  defined as indicated

$$A_1 = \frac{\mathbf{S} - \mathbf{S}_0}{\lambda} \cdot \mathbf{a}_1 = h_1 = h,$$

$$A_2 = \frac{\mathbf{S} - \mathbf{S}_0}{\lambda} \cdot \mathbf{a}_2 = h_2 = k,$$

$$A_3 = \frac{\mathbf{S} - \mathbf{S}_0}{\lambda} \cdot \mathbf{a}_3 = h_3 = l \pm \phi/M_3.$$

$\mathbf{S}_0$  and  $\mathbf{S}$  are unit vectors in the direction of the incident and diffracted beams, respectively,  $\lambda$  is the wavelength of the radiation, and  $\phi$  is the phase shift in the diffracted beam arising from the insertion of the domain boundary. The unit cell-structure factor for the  $(00.l)$  reflection is given normally by Eq. (1). If we introduce an antiphase boundary, there will be a shift or displacement  $\delta$  of one cell with respect to its neighbor across the boundary of

$$\delta = \mathbf{a}_1/3 + \mathbf{a}_2/6 + \mathbf{a}_3/6, \quad (3)$$

with an associated phase shift  $\phi$  given approximately by

$$\phi = \frac{\mathbf{S} - \mathbf{S}_0}{\lambda} \cdot \delta = h/3 + k/6 + l/6 \quad (4)$$

and this becomes  $\phi = l/6$  for the  $(00.l)$  reflection. The structure factor of the phase-shifted cell may then be expressed as

$$F'(00.l) = F(00.l) \exp[\pi il/3] \quad (5)$$

and  $F'(00.l) = -24p$  for  $l = 6n \pm 3$ . For antiphase boundaries normal to  $[00.l]$ ,  $M_1$  and  $M_2$  are both large numbers, and the intensity of the  $(00.l)$  reflection is given approximately by

$$I(00.l) = K p^2 M_1^2 M_2^2 \frac{\sin^2 \pi M_3 A_3}{\sin^2 \pi A_3} \times \frac{\sin^2 \pi N_3 M_3 (1 - \phi/M_3)}{\sin^2 \pi M_3 (1 - \phi/M_3)}, \quad (6)$$

where  $K$  involves other intensity factors which are not important here. The maxima in  $I(00.l)$  now do not come exactly at  $(00.l)$ , but are displaced along  $[00.l]$  by

an amount

$$\Delta h_3 = \pm \phi/M_3 = \pm 1/6M_3. \quad (7)$$

Thus, the  $(00.3)$  reflection now includes two satellites whose separation in reciprocal space along  $[00.l]$  is  $2\Delta h_3 = (3M_3)^{-1}$ . From the spacing of these satellites the size of the one-dimensional antiphase domain can be obtained.

Let us also consider the intensity and breadth of these new satellite reflections. It should be noted that the center portion does not disappear completely, because at  $A_3 = l$ , the first of the two functions in Eq. (6) has a very weak maximum given by

$$M_3^2 = \frac{\sin^2 \pi M_3 A_3}{\sin^2 \pi A_3} \Big|_{A_3=l}$$

and a full breadth given by  $1/M_3$ . The second function in Eq. (6) goes to a maximum at  $l \pm (6M_3)^{-1}$  with an intensity proportional to

$$N_3^2 = \frac{\sin^2 \pi N_3 M_3 (1 - \phi/M_3)}{\sin^2 \pi M_3 (1 - \phi/M_3)} \Big|_{A_3=l \pm 1/6M_3}$$

and a width of  $1/N_3 M_3$ . If the crystal has a fully-ordered domain structure along  $[00.l]$ ,  $N_3$  may correspond to thousands of domains, whereas  $M_3$  is characteristically about 5–10 unit cells. Under these conditions, the central peak is negligibly small. If  $N_3$  is comparable to  $M_3$ , then both the central and the satellite peaks are comparable in intensity. For  $N_3 = M_3$ , corresponding to the case where the number of domains maintaining coherence is equal to the number of cells within a domain, the satellite and central peaks have the same intensity. The widths are in the ratio, satellite/central =  $1/N_3$ . If the periodicity is also altered by allowing a range of integral values for  $M_3$  and  $N_3$ , the three peaks become further broadened by the superposition. Since instrumental broadening is also present, it is thus possible, for the case of small domains in the disordered state, to smear out the entire satellite pattern into a broad  $[00.l]$  streak.

Since satellites are observed above  $T_N$ , a fine domain network must exist with reasonably well-defined average values of  $M_3$  and  $N_3$ . Since  $M_3$  and  $N_3$  are small numbers, the total diffuse intensity is not produced by a single interference function  $G^2$  and must include, as suggested earlier, the superposition of these functions from all such small regions in the crystal. As  $M_3$  and  $N_3$  decrease, it may also be meaningful to adopt a random antiphase model<sup>8</sup> in which the entire disordered structure is built up of randomly-spaced tiny domains of average size  $M_3$ . However, this model yields the limiting case of directional broadening with no satellites. It thus represents a way of superposing the previous solutions for  $N_3 = 1$  with a well-defined average  $M_3$ .

We now apply the antiphase model to the scattering in Figs. 1 and 2. From the mean satellite positions we obtain an average domain size of about three hexagonal unit cells, or  $46 \text{ \AA} \pm 5 \text{ \AA}$  at  $133^\circ\text{K}$  and about  $61 \text{ \AA} \pm 10 \text{ \AA}$  at  $298^\circ\text{K}$ . The weaker and more diffuse intensities at  $298^\circ\text{K}$ , which suggest more randomness in spin direction, contain a much greater uncertainty than the  $133^\circ\text{K}$  results. We believe that the  $133^\circ\text{K}$  results are more reliable and attach no significance to the fact that the apparent domain size at room temperature is larger than at  $133^\circ\text{K}$ . Since the satellite and central peak intensities are about equal, we may assume that the coherency length of a single sequence is about three domains, indicating that these domains of local spin layering do not repeat very often. It is possible to have a disordered domain structure consisting of many small nuclei of order along  $[00.l]$ , but interrupted by material with either a random-spin structure or, equivalently, a much more disorganized domain network. The model thus becomes essentially one of small, reasonable well-ordered, antiphase nuclei embedded in a nearly random matrix.

There are other causes of satellite or side-band spectra in solid alloys from which we might deduce, by analogy, alternate explanations for the present results. For instance, among the variety of clustering alloys there is one particular example in which a periodic modulation of alloy composition is accompanied by a periodic modulation of lattice constant, but no modulation in scattering power. This alloy, CuNiFe, has almost identical x-ray scattering factors for all the constituents. Using the model for CuNiFe developed by Hargreaves<sup>11</sup> to explain the side-band reflections, we can treat MnO as an  $[00.l]$  layered structure characterized by ordered layers, followed by disordered layers, in a periodic sequence in which only the  $[00.l]$  planar spacing changes as we proceed along the array. Satellites are predicted for this model at  $\pm(1/6M_3)$ , where  $M_3$  is again the number of unit cells in one domain of the sequence. The intensity of the central  $(00.l)$  spot is now given by the zeroth-order Bessel function,  $I_0 = J_0(8lM_3\epsilon/\pi d)$ , and the intensity of the first-order satellite by,  $I_1 = J_1(8lM_3\epsilon/\pi d)$ . Here,  $d$  is the average spacing between neighboring planes of Mn ions and  $\epsilon$  is the difference in interplanar spacing between the ordered and disordered regions. For the  $(00.3)$  reflection at  $133^\circ\text{K}$ ,  $M_3 \approx 3$ ,

$\epsilon = 0.015 \text{ \AA}$ ,  $d = 2.55 \text{ \AA}$  and the ratio  $I_1/I_0 = 0.067$ . That is, the side bands should be much weaker than the central spot. The observed intensity ratio is about  $I_1/I_0 = 0.9$ , and it appears that a model involving only a modulation of interplanar spacing does not predict sidebands of sufficient relative intensity.

There is also the possibility of composition fluctuations in the MnO crystal on a scale of about  $50 \text{ \AA}$ . Such fluctuations are not unreasonable, but satellites would be produced in the vicinity of the nuclear diffraction peaks and the satellite intensity would not be a function of temperature. Satellites were absent in the regions surrounding the nuclear peaks and we conclude, therefore, that antiphase domains with a relatively well-defined size are present in MnO above the Néel temperature. It is also possible to explain the other observations discussed earlier. The shifting of both the central diffusive peak and the mean satellite position to higher angles probably has its origin in the  $(111)$  planar spacing contraction of the locally ordered regions. If we assume that these regions have the lattice parameter of fully ordered MnO, while the  $(111)$  position is defined by the disordered magnetic cell parameter, an outward shift of  $0.026 \text{ rlu}$  would be expected at  $133^\circ\text{K}$ . This is reasonably close to the observed value of  $0.043 \text{ rlu}$ . The ordered MnO lattice becomes rhombohedral with a rhombohedral angle of  $90^\circ 30'$ . A cumulative tilt of  $(111)$  planes over four repeating domains would account for the observed 2-deg tilt of the axis relative to the average  $\langle 111 \rangle$  direction in the matrix, but it is not clear why such a regular cumulative tilt should arise.

We believe that the domain analysis of these satellite patterns is appropriate and that the estimated sizes are quite reasonable. A much more extensive study is currently underway in which we hope to obtain a more exact set of correlation functions which include a specification of spin direction, spin coupling, and correlation range, and the variation of these parameters with temperatures.

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<sup>11</sup> M. E. Hargreaves, Acta Cryst. 4, 301 (1951).