Direct Evidence for Triple-Q Spin-Density Wave in fcc Antiferromagnetic Mn-Ni Alloy

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It is shown by a brief calculation that the method of anisotropy measurement of γ-ray emission from spin-polarized nuclei is a useful tool to investigate multiple-spin-modulation waves which have been suggested to exist in several kinds of materials but have not yet been proven by definitive experimental evidence. An application of this method is made for a fcc antiferromagnetic Mn-Ni alloy and the first direct experimental evidence for a triple-Q spin-density wave is given.

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In several fcc transition-metal alloys (MnNi,[1] MnFe,[2] MnPd,[3] etc.) antiferromagnetic order of the first kind has been predicted to exist in neutron-diffraction experiments. The magnetic-moment configuration of this order is described as a commensurate collinear spin-density wave with a longitudinal spin polarization, that is,

$$\mu_j(\mathbf{r}) = \mu \frac{\mathbf{Q}_j}{|\mathbf{Q}_j|} \exp(i\mathbf{Q}_j \cdot \mathbf{r}) + \text{c.c.}, \quad j = x,y,z, \qquad (1)$$

where the $\mathbf{Q}_j$'s are magnetic wave vectors, $\mathbf{Q}_x = (2\pi/a)(100)$, $\mathbf{Q}_y = (2\pi/a)(010)$, and $\mathbf{Q}_z = (2\pi/a)(001)$, and a is the lattice constant [see Fig. 1(a)]. We define the "single-Q state' $|\mathbf{Q}_j\rangle$ as a quantum state in which $\mu_j(\mathbf{r})$, which involves only one wave vector, stands in the crystal. In a cubic crystal these three single-Q states are crystallographically equivalent to each other. If these states are degenerate, magnetic domains will be formed in the crystal for each $|\mathbf{Q}_j\rangle$. Several theoretical calculations[4-6] were made to examine the stability of the single-Q state and predicted that under certain circumstances the single-Q state becomes unstable relative to the "multiple-Q state,"

$$|\mathbf{Q}_M\rangle = \frac{1}{\sqrt{V}} \{\sqrt{v_x}\,|\mathbf{Q}_x\rangle + \sqrt{v_y}\,|\mathbf{Q}_y\rangle + \sqrt{v_z}\,|\mathbf{Q}_z\rangle\}, \qquad (2)$$

in which the magnetic-moment configuration,

$$\mu_M(\mathbf{r}) = \frac{1}{\sqrt{V}} \{\sqrt{v_x}\mu_x(\mathbf{r}) + \sqrt{v_y}\mu_y(\mathbf{r}) + \sqrt{v_z}\mu_z(\mathbf{r})\}, \qquad (3)$$

is noncollinear as is shown in Fig. 1(b). In Eqs. (2) and (3), the v_j's are constants and $V = v_x + v_y + v_z$ [the spin configuration shown in Fig. 1(b) corresponds to the case $v_x = v_y = v_z$]. For example, Jo considered[5] the effect of the four-spin interaction in fcc transition-metal alloys on the stability of the multiple-Q state and predicted that the triple-Q state will occur when the 3D electron band is nearly half-filled.

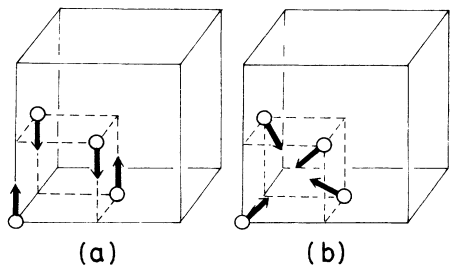

FIG. 1. (a) The spin configuration of single-Q and (b) triple-Q structures on a fcc lattice based on a longitudinally polarized antiferromagnetic modulation with propagation vectors $(2\pi/a)\langle 100\rangle$. Only the four sublattice moments are shown. In (b), each spin points in a $\langle 111\rangle$ direction.

Experimentally, it is very difficult to prove the existence of the multiple-Q state. For example, let us consider a crystal in which the volume of each domain of the single-Q state $|\mathbf{Q}_j\rangle$ is v_j. The pattern of the neutron diffraction from such a crystal is completely the same as that from a crystal in which a multiple-Q state with a form of Eq. (2) is realized: In either case the intensity of the Bragg reflections at $\boldsymbol{\kappa} = \mathbf{G} \pm \mathbf{Q}_j$ are proportional to v_j/V, where $\boldsymbol{\kappa}$ and $\mathbf{G}$ are the scattering vector and the reciprocal lattice vector, respectively. This fact was first pointed out by Kouvel and Kasper.[7] Since then a lot of materials, including rare-earth compounds,[8,9] have been discovered as candidates for a multiple-Q antiferromagnet, but there has been no experimental evidence up to the present which could prove definitively the existence of the multiple-Q state. In this Letter we demonstrate by calculation that the method of anisotropy measurement of γ-ray emission from spin-polarized nuclei is useful to distinguish between the multiple-Q state and the single-Q state, and show the result of an application of

the method for an antiferromagnetic fcc MnNi alloy.

Since our current interest is in manganese alloys, we describe here only the properties of the γ-ray emission from a ^{54}Mn isotope. For other elements the same arguments are possible if a suitable isotope is available. In Fig. 2 we show the decay scheme of a ^{54}Mn nucleus. The emission probability of the γ ray has an anisotropy which depends on the initial nuclear-spin direction and on the characteristics of the decay.[11] In the case of ^{54}Mn the emission is of E_2 type (electric quadrupole emission) and the normalized intensity pattern of the emission has the form:

$$W(\theta)=1+\sum_{k=2,4} B_k U_k F_k P_k(\cos\theta), \tag{4}$$

where θ is the angle between the direction of the emission and the spin quantizing axis. The B_k's are the kth moments of the nuclear-spin direction which is determined by the magnitude of the spin polarization, i.e., in the present case, by the magnitude of the hyperfine field and the temperature. The U_k's are constants describing the effect of the transitions preceding the one detected, the F_k's are angular-momentum coupling constants for the emission being observed, and the P_k's are normalized Legendre polynomials,

$$\begin{aligned} P_2(\cos\theta)&=\tfrac{1}{2}(3\cos^2\theta-1),\\ P_4(\cos\theta)&=\tfrac{1}{8}(35\cos^4\theta-30\cos^2\theta+3). \end{aligned} \tag{5}$$

The values of the products U_2F_2 and U_4F_4 for ^{54}Mn are given in Ref. 11 and are -0.49486 and -0.44669, respectively, and the values of B_2 and B_4 are also tabulated there as functions of Δ/k_BT, where Δ is the magnitude of the Zeeman splitting of the nuclear sublevels. One can see the pattern of the angular emission probability of γ rays from a ^{60}Co isotope in Lounasmaa.[12] The emission pattern of ^{54}Mn is similar to that of ^{60}Co: At 0 K, there is no emission probability along the spin axis, while the emission is spherically isotropic when the spin has no polarization. Thus, the characteristics of the anisotropic γ-ray emission from ^{54}Mn is completely known and it is straightforward to derive the emission pattern from the antiferromagnetically aligned nuclei in a crystal. The P_2 term in Eq. (4) has a vanishing contribution to the emission intensity when the nuclei have a cubic configuration. In this case the anisotropy of emission originates from the $\cos^4\theta$ term in P_4.

According to Eq. (2), one can construct, in principle, an infinite number of multiple-**Q** states using an infinite number of the combination (v_x,v_y,v_z). However, we hereafter consider only the case of $v_x=v_y=v_z$ because this was the case for our sample used in the experiment which is described later. The condition $v_x=v_y=v_z=V/3$ corresponds to either case of the single-**Q** phase with an equal population for all the $|\mathbf{Q}_j\rangle$ domains or the triple-**Q** phase with $|\mathbf{Q}_M\rangle=(1/\sqrt{3})\{|\mathbf{Q}_x\rangle+|\mathbf{Q}_y\rangle+|\mathbf{Q}_z\rangle\}$. In Fig. 1, one can see that the set of the directions of the spins which are realized in the crystal in the case of $v_x=v_y=v_z$ is $\pm\{[100],[010],[001]\}$ for the single-**Q** phase with three equally populated domains and $\{[111],1\bar{1}\bar{1}],[\bar{1}1\bar{1}],[\bar{1}\bar{1}1]\}$ for the triple-**Q** phase. In each set the probabilities of having each direction are equal to each other. Then, with α_1, α_2, and α_3 the direction cosines of the direction of observation, the normalized intensity patterns from the respective set of spin directions are represented as

$$\begin{aligned} W_s(\alpha_1\alpha_2\alpha_3)&=(1+\tfrac{7}{12}B_4U_4F_4)-\tfrac{35}{12}B_4U_4F_4(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2),\\ W_t(\alpha_1\alpha_2\alpha_3)&=(1-\tfrac{7}{18}B_4U_4F_4)+\tfrac{35}{18}B_4U_4F_4(\alpha_1^2\alpha_2^2+\alpha_2^2\alpha_3^2+\alpha_3^2\alpha_1^2). \end{aligned} \tag{6}$$

In Eqs. (6), it is essential that the anisotropy terms in W_s and W_t have opposite signs to one another. Because of this fact, one can distinguish between the single-**Q** phase and the triple-**Q** phase simply by a qualitative analysis of experimental data. In Fig. 3, the calculated intensity of emission in the (001) plane between the [100] and [010] directions is shown for several temperatures. One can see that the emission intensity has a maximum in the [110] direction and a minimum in the [100] direction for the single-**Q** structure and vice versa for the triple-**Q** structure.

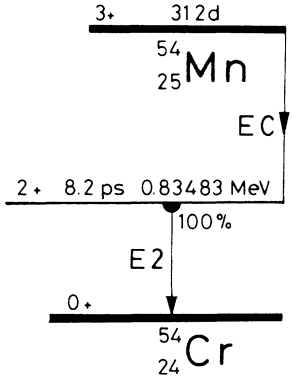

FIG. 2. The decay scheme of a ^{54}Mn isotope from Ref. 10.

The Zeeman splitting of the nuclear sublevels, Δ, is caused by the hyperfine field from the polarized electronic moment and in the case of ^{54}Mn, Δ is given as[11]

$$\Delta/k_B=g_N\mu_N H_{hf}/k_B=0.0403H_{hf}\ (\mathrm{mK}), \tag{7}$$

where g_N is the nuclear g factor, μ_N is the nuclear magneton, and the hyperfine field, H_{hf}, is measured in units of kilo-oersteds. The hyperfine fields at Mn nuclei in the Mn-Ni alloy system have been measured at low temperature by Takenaka and Asayama[13] using NMR. Reflecting the chemical disorder of the alloy, H_{hf} has a

broad spectrum between 60 and 190 kOe with a center of gravity at 120 kOe. The splitting of the nuclear sublevels corresponding to a H_{hf} of 120 kOe is $\Delta/k_B = 4.8$ mK. Thus, it is necessary to cool the sample much below 5 mK to reach a fully polarized state of the nuclear spins.

We prepared a small ($10 \times 5 \times 1$ mm^3) single crystal of ^{54}Mn-doped $Mn_{72}Ni_{28}$ alloy. Doping of ^{54}Mn was made by following the method used by Berglund *et al.*[11] The γ-ray radioactivity of the sample was about 1 μCi. This magnitude of radioactivity causes no serious self-heating problem in the sample even at very low temperature.[12] From the magnetic phase diagram given by Honda, Tanji, and Nakagawa,[14] the Néel temperature of this sample is estimated to be about 420 K. The crystalline mosaic spread of this crystal was examined by neutron diffraction and was less than 0.6°, which is small enough for the present purpose. The observed intensities of the antiferromagnetic Bragg reflections of neutrons corresponding to each $|\mathbf{Q}_j\rangle$ were all the same, which indicates $v_x = v_y = v_z$.

The sample was soldered to a copper sample holder with indium solder. Then, the sample holder was screwed onto the copper-nuclear-demagnetization state (ND stage) of a ^{3}He/^{4}He dilution refrigerator. With use of a small coil, a small magnetic field (300 Oe) was applied to the sample in order to improve the thermal conductivity of the solder by breaking its superconductivity. This magnitude of external field gives no effect on the distribution of the antiferromagnetic domains which have already been formed at the Néel temperature. A platinum-NMR thermometer and a ^{3}He-melting pressure thermometer were used to monitor the temperature of the ND stage. The γ photons were counted with use of a pure-germanium solid-state detector.

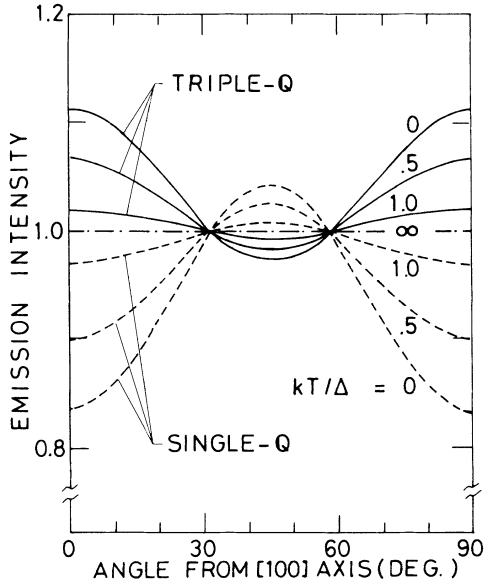

FIG. 3. The calculated intensity pattern of emission from a single-Q and a triple-Q antiferromagnet at different temperatures. The direction of observation is between [100] and [010] in the (001) plane.

Because of the limiting measuring time, it was difficult to count at many different directions. We, therefore, fixed the detector angle at [100] direction, where the largest change of the emission intensity with changing temperature is expected, and compared the intensities measured both at the lowest temperatures attainable (around 1 mK) and at high enough temperatures (around 20 mK) to depolarize the nuclear spins. Measurements were repeated at the high and the low temperatures to check the reproducibility of the data.

In Table I are shown the results of the measurements. The data II and IV were all taken after a measurement at high temperature and are normalized to the high-temperature counts. The actual numbers of counts were typically 2×10^5. A correction was made for the effect of the natural decay of the ^{54}Mn isotopes during the measuring time. In Table I, one can see that the largest change of the emission intensity when the sample was cooled is as much as 4.6%. This change is large compared to the statistical error.

The result which is obtained here indicates that the triple-Q configuration of the electronic moments is realized in this material. At 0.2 mK, the nuclear-spin polarization in the sample is expected to be almost complete. Then Fig. 3 predicts a change of the emission of about 12% at this temperature for a perfect triple-Q spin configuration. The observed change of 4.6% is considerably less than this value. Several speculations are possible for this disagreement: It is so difficult to measure directly the temperature of the sample itself that we only monitored the temperature of the ND stage. There may have been a temperature difference between the sample and the ND stage. A temperature difference of about 3 mK can explain this disagreement. Also, the calculation for Fig. 3 is based on a perfect triple-Q spin configuration. But, since our sample is a disordered alloy it is very likely that some Mn moments, depending on their near-neighbor atomic configuration, deviate from the right directions of the triple-Q alignment. Such a devia-

TABLE I. Emission intensity measured in the [100] direction at high and low temperatures. The designations I–IV indicate the order of the repeated measurements. The intensities are normalized to the warm counts, I.

	Temperature (mK)	Emission intensity	Statistical error (%)
I	18.0	100.00	0.33
III	17.0	100.10	0.33
II	1.5	102.22	0.37
IV	0.2	104.57	0.37

tion should cause a reduction of the anisotropy of the emission.

In any case, however, in the present experimental condition, there is no other reason for the increase of emission in the [100] direction with decreasing temperature than the alignment of the nuclear spins toward the triple-Q configuration. We therefore regard the present result to be unequivocal evidence for the existence of a triple-Q spin-density wave in this material.

fcc Fe-Mn alloy with a Mn concentration of 20%–65% is also a candidate for triple-Q antiferromagnetism.[2] Kennedy and Hicks[15] have recently made a Mössbauer transmission experiment of a single-crystal $Fe_{56}Mn_{44}$ alloy in a strong magnetic field and deduced the existence of a triple-Q phase in this material. On the other hand, Bisanti, Mazzone, and Sacchetti[16] have predicted the existence of a single-Q phase in $Fe_{66}Mn_{34}$ alloy by analyzing the results of spin-wave measurement by neutron scattering. Our method should be able to give a definitive answer to these contradictory results.

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