Solid State Communications, Vol. 11, pp. 391–394, 1972. Pergamon Press. Printed in Great Britain

INELASTIC NEUTRON SCATTERING STUDY OF SPIN WAVES IN MnO

M. Kohgi, Y. Ishikawa and Y. Endoh

Department of Physics, Tohoku University, Sendai 980, Japan

(*Received* 6 *May* 1972 *by* *T. Nagamiya*)

The dispersion relation $E(q)$ for spin waves in MnO has been measured at 4.2°K by neutron inelastic scattering. The isotropic exchange integrals as well as the anisotropy constants have been determined by fitting the Hamiltonian to the data. It is found that the exchange striction plays main role in the anomaly in the magnetic interactions in MnO and the biquadratic exchange interaction $j_1(S_1 \cdot S_2)^2$ is almost absent ($j_1/J_1 \lesssim 0.002$).

MANGANESE mono-oxide, MnO, is a typical antiferromagnet with the f.c.c. Type II structure ($T_N = 117$°K). The magnetic interactions in this substance have provoked the attention of many investigators, because the presence of the biquadratic interaction $j(S_1S_2)^2$ has been suggested in connection with the ESR spectrum of Mn^{2+} pairs in MgO[1] and the anomalous temperature dependence of sublattice magnetization in MnO.[2] We have measured the spin wave dispersion relations in MnO by neutron inelastic scattering technique to determine the exchange and anisotropy parameters accurately.

A single crystal of MnO, about 3 cm³ in volume,[3] was used as the sample, which had been annealed for 3 hours at 1450°C in argon atmosphere in order to eliminate the internal strain. The measurements were carried out on a triple axis spectrometer of Tohoku University installed at JRR-2* at 4.2°K and higher temperatures using both constant-Q and constant-E techniques. Incident neutron energies of 30 and 50 meV were used. The spin waves were observed along three crystallographic directions [111], [001], and $[\bar{1}\bar{1}1]$ around the magnetic Bragg scattering point (111). These indices are referred to the reciprocal lattice of a pseudo-cubic cell† of side $2a$, a being the lattice parameter of the chemical unit cell.

There are some complications because of the presence of four types of domains (T-domains) in the Type II antiferromagnetic structure corresponding to the four {111} planes of ferromagnetic ordering. Since the spin waves have two modes for each wave vector q in a domain, eight branches should in general be observed for one crystallographic direction in a multi-domain crystal. The specimen was found to contain the four domains in nearly equal proportion, but the spin waves in different domains could be distinguished by symmetry consideration. Two modes of each domain could not be separated except near the magnetic Brillouin zone center (111).

The spin wave dispersion relations measured at 4.2°K are shown in Fig. 1. They have not yet been corrected for the instrumental resolution effects. The wave vector of a spin wave is expressed, for example, by $q = \pi/a\,[\xi,\xi,\xi]$. The result of antiferromagnetic resonance (AMFR) at 2°K[4] as well as our result at about 20°K are also included for reference.

* Japan Research Reactor 2 at JAERI.

† MnO shows small rhombohedral distortion along [111] in antiferromagnetic phase.

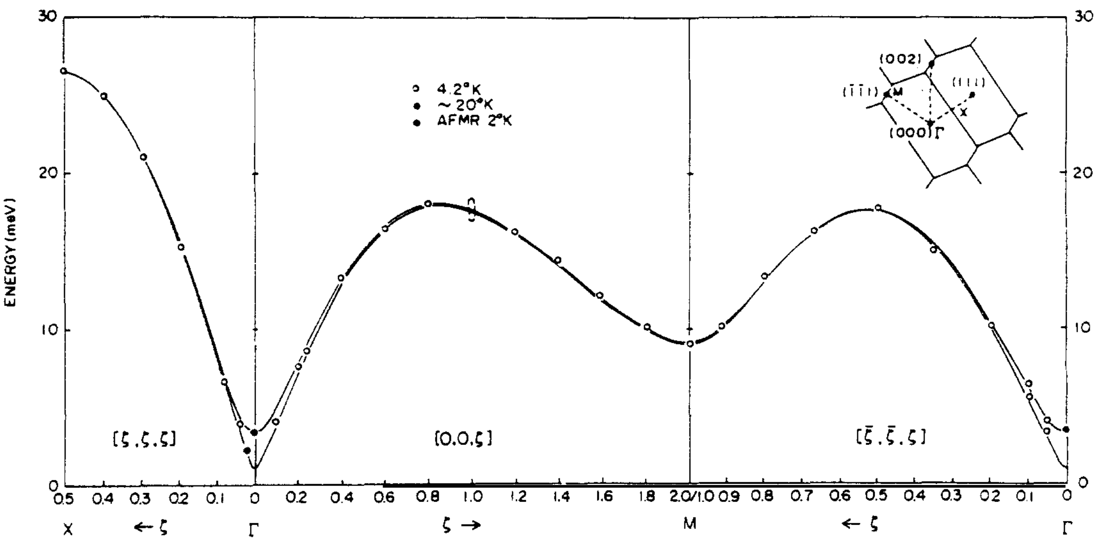

FIG. 1. Dispersion relation $E(q)$ of the spin waves in MnO measured along [111], [001] and [$\bar{1}\bar{1}$1].

The Hamiltonian of MnO is assumed as

$$H = \sum_{nn}^{a} J_1^{+} S_i S_j + \sum_{nn}^{p} J_1^{-} S_i S_j + \sum_{nnn} J_2 S_i S_j + \sum_{nn} j_1 (S_i S_j)^2 + \sum_{nnn} j_2 (S_i S_j)^2 + \sum_{n} D_1 (S_{iz})^2 + \sum_{n} D_2 (S_{iy})^2 .$$

The right hand side of the first line of the above expression represents the bilinear exchange interactions involving the effects of the exchange striction which cause the thombohedral distortion below T_N and modify the nearest neighbour (nn) exchange integral J_1 to J_1^{+} for antiparallel pairs and to J_1^{-} for parallel pairs. The next nearest neighbour (nnn) exchange integral J_2 suffers only from uniform deformation by this effect. Σ^a and Σ^p denote summation over antiparallel pairs and parallel pairs, respectively. D_1 and D_2 terms are approximate forms of the out of plane and in plane anisotropy energies, respectively, which are thought to be due to the dipolar and higher order interactions.

The Hamiltonian can be solved in the free spin wave approximation by the standard method to give two modes of spin waves for each wave vector $q = (q_x, q_y, q_z)$, whose energies are

$$\begin{aligned} E_1(q) = S[\{2D_1 &+ (\gamma_{10} - \gamma_{1q}) J_1^{+'} + (\gamma_{20} - \gamma_{2q}) J_1^{-'} + (\gamma_{30} - \gamma_{3q}) J_2' \} \\ &\times \{2D_2 + (\gamma_{10} - \gamma_{1q}) J_1^{+'} + (\gamma_{20} - \gamma_{2q}) J_1^{-'} + (\gamma_{30} - \gamma_{3q}) J_2' \}]^{1/2} , \end{aligned}$$

where

$$J_1^{+'} = J_1^{+} - 2j_1 S^2 = J_1 + \Delta J,$$
$$J_1^{-'} = J_1^{-} + 2j_1 S^2 = J_1 - \Delta J,$$
$$J_2' = J_2 - 2j_2 S^2,$$
$$\gamma_{1q} = 2\left[\cos\frac{q_x + q_y}{2}a + \cos\frac{q_y + q_z}{2}a + \cos\frac{q_z + q_x}{2}a\right],$$
$$\gamma_{2q} = 2\left[\cos\frac{q_x - q_y}{2}a + \cos\frac{q_y - q_z}{2}a + \cos\frac{q_z - q_x}{2}a\right],$$
$$\gamma_{3q} = 2[\cos q_x a + \cos q_y a + \cos q_z a],$$

and $E_2(q)$ which is given by the same expression as $E_1(q)$ with the terms D_1 and D_2 interchanged. The neutron cross section of the two modes are not equal and are rather complex functions of q; for example $E_2(q)$ is not observed in the [111] direction.

The best-fit procedure has been applied to the observed spin waves. The solid lines in

Fig. 1 are the calculated curves of $E_1(q)$ and $E_2(q)$ using following parameters:

$$J_1 = 0.77 \pm 0.02,\ J_2' = 0.89 \pm 0.02,$$
$$\Delta J = 0.09 \pm 0.01$$
$$D_1 = 0.043 \pm 0.002,\ D_2 \sim D_1 \times 10^{-1}$$

all in units of meV. The agreement between these curves and the observation is satisfactory within experimental errors. It is noted that the spin wave energy of 9 meV at M point in the figure cannot be explained without assuming the presence of the ΔJ terms. The exchange interactions between further neighbours than the second seem to be negligible. The obtained parameters are distinctly different from the values determined by the early spin wave study,[5] but are nearly in good agreement with those deduced by Lines and Jones from static measurements.[6] This suggests that the parameters determined at 4.2°K by the spin wave theory explain consistently the static magnetic properties.

The contribution of the exchange striction to ΔJ, ΔJ_s, is easily estimated considering that the rhombohedral distortion is caused by the exchange striction.[6,7] The distortion makes small difference between the separation of nn antiparallel and parallel pairs, which results in

$$J_1 = J_1\left(1 \pm \frac{1}{2}\epsilon\Delta\right) = J_1 \pm \Delta J_s,$$

where $\epsilon = d_1(\partial J_1/\partial r)/J_1$, d_1 the average separation of nn pairs, and Δ the change of the corner angle of the cubic lattice by the distortion. The equilibrium value of Δ is determined by the balance condition between the elastic and exchange energies to be $\Delta_{eq} = Nz_1J_1\epsilon(\bar{S})^2/4C$, where z_1, N, $(S)^2$ and C are the number of spins in the system, the thermal average of $(S_iS_j)^2$ over nn parallel pairs, and the elastic constant ($= 3C_{44}$) respectively. The contribution ΔJ_s ($= 1/2\ \epsilon J_1\Delta_{eq}$) was estimated by Lines and Jones using the relation between Δ_{eq} and ϵJ_1 mentioned above to be $\Delta J_s = 0.11$ meV. This value is almost the same as ΔJ determined by this work within the experimental errors. Therefore it may be concluded that the anomaly in the magnetic interactions in MnO is explained in terms of the exchange striction, in accordance with the conclusion of Lines and Jones.[6] The order of the biquadratic exchange interaction is estimated to be $j_1/J_1 \lesssim 0.002$.

The spin wave measurements at 60 and 77°K have shown that the spin wave energies decrease almost uniformly with increasing temperature throughout the whole branches (by about 1 meV at [0.5, 0.5, 0.5], [0, 0, 1] and [−0.5, −0.5, 0.5] at 60°K), except near [002] (the M point) where the decrease is remarkable. It is found that about half of the decrease at M can be explained in term of the temperature dependence of ΔJ_s which is proportional to $\Delta_{eq}{}^{1/2}$.

The origin of the nearest neighbour interaction between Mn^{2+} ions is of particular interest, This has nearly the same order of magnitude as the 180° superexchange interaction and is strongly distance-dependent ($[\partial J_1/\partial r] = 6.4$ meV/Å). A full report will be published elsewhere.

Acknowledgements – The authors wish to thank Prof. K. Motizuki, Prof. E. Uchida and Prof. T. Yamada for helpful discussions. The assistance of Dr. K. Tajima with the experimental techniques is also greatly appreciated. This work was partially supported by the Grant-in-Aid for Scientific Research from Ministry of Education.

REFERENCES

1. HARIS E.A. and OWEN J., *Phys. Rev. Lett.* **11**, 9 (1963).
2. RODBELL D.S., JACOBS I.S. and OWEN J., *Phys. Rev. Lett.* **11**, 10 (1963); HUANG N.L. and ORBACH R., *Phys. Rev. Lett.* **12**, 275 (1964).
3. The crystal was purchased from Nakazumi crystal Co.
4. RICHARDS P.L., *J. appl. Phys.* **34**, 1237 (1963).

5. COLLINS M.F., Report on the *Int. Conf. on Magnetism*, Nottingham, (1964).

6. LINES M.E. and JONES E.D., *Phys. Rev.* **139**, A1313 (1965).

7. RODBELL D.S. and OWEN J., *J. appl. Phys.* **35**, 1002 (1964).

On a mesuré la relation de dispersion $E(q)$ des ondes de spins dans MnO à 4.2°K par la diffusion inélastique neutronique. Quelques paramètres des interactions d'échange ainsi que celles de l'anisotropie ont été déterminé à partir de ces résultats. On a trouvé que la striction d'échange joue un rôle essentiel dans l'anomalie des interactions dans MnO, ce qui suggère que l'interaction d'échange biquadratique $j_1(S_1 \cdot S_2)^2$ est presque null ($j_1/J_1 \leqslant 0.002$).