JOURNAL OF THE PHYSICAL SOCIETY OF JAPAN VOL. 17, SUPPLEMENT B-III, 1962
PROCEEDINGS OF INTERNATIONAL CONFERENCE ON MAGNETISM AND CRYSTALLOGRAPHY, 1961, VOL. III

An Investigation of Magnons in Franklinite by the Neutron Scattering Method*

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The concept of spin waves or magnons has an important place in the modern theory of magnetic phenomena. These quasi-particles reflect the collective behaviour of electronic spins strongly coupled by exchange forces in ferro-, ferri- or antiferromagnetics at temperatures sufficiently below their critical points. They carry momentum $\hbar q$ (q=wave vector) as well as energy $\hbar\omega$ (ω=angular frequency). The basic property of a spin wave is its dispersion relation, i.e., connection between ω and q.

Neutrons interact with spin lattices by magnetic forces and can undergo inelastic scattering with the creation or annihilation of magnons. During this process the conservation laws of energy and momentum must be fulfilled. In principle it gives a unique method for the investigation of magnons[1]. All that is needed is to measure the energy and momentum of neutrons before and after scattering on a given spin system. Unfortunately, relevant cross sections are of the order of a few milibarns per magnetic ion and unless very strong neutron beams are available some less direct methods have to be applied. Riste *et al.*[2,3] have obtained valuable results in the case of Fe_3O_4 and Fe_2O_3 using a white neutron beam without analysis of neutron energy.

For franklinite essentially the same technique was applied. The modification was that the white neutron beam was replaced by a monochromatic one (λ_0=1.41 Å). Experiments were performed on a large single crystal of natural origin. Its chemical composition could be satisfied by the formula: $Zn_{0.75}Mn_{0.38}Fe_{1.87}O_4$. The neutron diffraction study (to be published elsewhere) using a small fraction cut from the crystal supported the following cation distribution among spinel A and B sites: $(Zn_{0.75}Mn_{0.25})_A(Fe_{1.87}Mn_{0.13})_BO_4$ and agreed satisfactorily with the magnetization measurements although the net magnetic moment per "molecule" appeared to be lower than could be predicted by a simple Néel's model.

Since the critical point of the sample was about 230°K, experiments on magnon-neutron scattering were done at liquid air temperature. The crystal was contained inside a cryostat, mounted on the neutron spectrometer table, with two rotational degrees of freedom. Throughout the course of the experiments the [110] axis of the crystal was aligned vertically. Thus diffuse scattering around the (111) Bragg reflection could be studied. For this purpose, a monoenergetic beam of neutrons obtained by reflection from an aluminium crystal monochromator was directed on the crystal. With the applied collimators (in a horizontal beam hole of the WWRS-reactor and on the spectrometer arm in front of the BF_3 counter) the standard half width of the (111) Bragg peak of the sample amounted to 42′.

The measurements consisted in the scanning of the intensity of the diffuse scattering peak appearing in place of the (111) Bragg reflection when the crystal was shifted away from the Bragg reflection position θ_B by a known angle $\Delta\theta=\theta-\theta_B$ (we take $\Delta\theta$ as positive for the magnon creation side). The diffuse peaks were scanned in the horizontal plane by a high efficiency neutron counter with about 40′ nominal collimation placed inside heavy shielding on the spectrometer arm.

The observed broad peaks were proved to be of predominantly magnetic origin, as for sufficiently large $\Delta\theta$ they almost disappeared after heating the crystal to 200°C. A comparison of the magnetic and nuclear structure factors of (111) reflection revealed that magneto-vibrational scattering at liquid air temperature should be much smaller than magnon scattering. For small $\Delta\theta$ there was observed

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a relatively strong admixture of Bragg scattering caused by the mosaic spread of the domains or misaligned crystallites of the sample. However, this scattering was restricted to the centre of the diffuse peak only, which is one of the advantages of the monochromatic beam technique. The wings of the diffuse peaks were (or should be) free from Bragg scattering contamination as well as magneto-vibrational or nuclear vibrational scattering. Hence at first the total widths of the diffuse peaks were taken into consideration. Since the wings of the diffuse peaks merged comparatively smoothly into the background it was necessary to estimate the width, to some extent arbitrarily, finding the intersections of extrapolated slope-lines with background level. For rough instrumental correction the half width of the Bragg peak (42′) was subtracted from each of the apparent total widths. The half of the total width ($\frac{1}{2}\Gamma_{\mathrm{tot}}$) of the diffuse peaks so corrected could be compared with theoretical calculations based on a quadratic dispersion relation for magnons ($\omega=(\hbar/2m_0)\alpha q^2$, m_0=neutron mass). As may be seen from Fig. 1 the agreement is satisfactory for the dispersion constant $\alpha\approx40$.

The examination of the shapes of the diffuse peaks in comparison with those theoretically predicted one supports a rather higher value of α, namely $\alpha=50\pm10$. Theoretically predicted shapes have been obtained

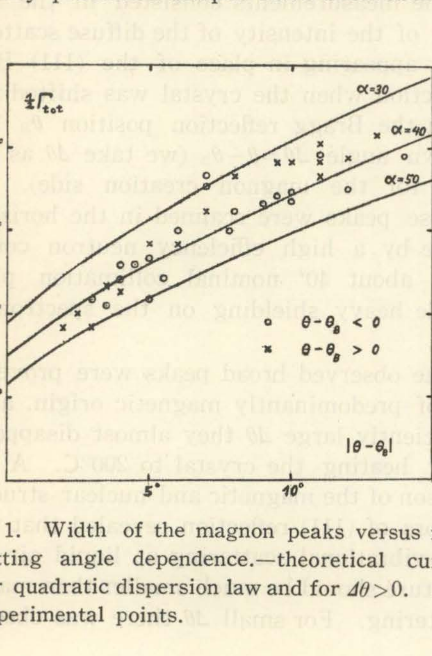

Fig. 1. Width of the magnon peaks versus missetting angle dependence.—theoretical curves for quadratic dispersion law and for $\Delta\theta>0$. ×○ experimental points.

in the usual manner by numerical integration over the scattering surface taking into account the density of magnon states, population factors given by B.-E. statistics and instrumental resolution represented by the Bragg peak curve. The difficulty is that the peaks have not exactly the calculated shapes, see Fig. 2. One can suspect[4] that the lifetimes

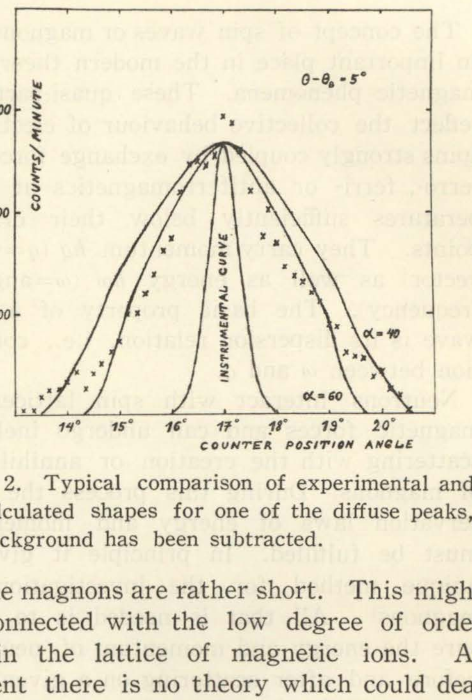

Fig. 2. Typical comparison of experimental and calculated shapes for one of the diffuse peaks, Background has been subtracted.

of the magnons are rather short. This might be connected with the low degree of order within the lattice of magnetic ions. At present there is no theory which could deal with such cases by the lack of translational order. However, from the general point of view one might expect rather quadratic dispersion relation and a relatively small value of α, which was in fact observed.

Up to now, neutron-magnon scattering has been studied only in a few magnetic substances all with high critical points and comparatively good order: Fe_3O_4, $T_C=855°K$, $\alpha\approx225$[2),5)]; Fe, $T_C=1043°K$, $\alpha\approx135$[6)]; $Co_{0.92}Fe_{0.08}$, $T_C=1300°K$, $\alpha\approx180$[7)]. For this reason, in the case of franklinite it would be very interesting to obtain precise data by the more direct method of Sinclair and Brockhouse[7)] in order to look for the lifetimes of the magnons.

References

1 R. J. Elliott and R. D. Lowde: Proc. Roy. Soc. **A230** (1955) 46.
2 T. Riste, K. Blinowski and J. Janik: J. Phys. Chem. Solids **9** (1959) 153.

3 T. Riste and A. Wanic: J. Phys. Chem. Solids **17** (1961) 318.
4 R. J. Elliot and H. Stern: I.A.E.A. Symposium on Inelastic Scattering of Neutrons in Solids and Liquids, Vienna, 1960.
5 B. N. Brockhouse: Phys. Rev. **106** (1957) 859.
6 R. D. Lowde and N. Umakantha: Phys. Rev. Letters **4** (1960) 452.
7 R. N. Sinclair and B. N. Brockhouse: Phys. Rev. **120** (1960) 1638.