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U.S.-JAPAN COOPERATIVE PROGRAM ON NEUTRON SCATTERING

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U.S.-JAPAN COOPERATIVE PROGRAM ON NEUTRON SCATTERING

ABSTRACT

The U.S.-Japan Cooperative Program on Neutron Scattering was implemented through arrangements by the United States Department of Energy with the Science and Technology Agency (STA) and the Ministry of Science, Education, and Culture (Monbusho) of Japan. It involves research collaboration in neutron scattering by Japanese scientists with scientists at Oak Ridge National Laboratory (ORNL) and Brookhaven National Laboratory (BNL) and the construction of new neutron scattering equipment at both laboratories with funds provided by the Japanese government. The United States provides neutrons in exchange for the new equipment, and other costs of the program are equally shared by the two countries. The assignments of Japanese scientists to ORNL and BNL vary in length, but they correspond to about two person years annually at each laboratory. An equal number of U.S. scientists also participate in the research program.

The main research collaboration is centered around the new equipment provided by the Japanese, but other facilities are utilized when they are needed. The new equipment includes a new type of wide-angle diffractometer and equipment for maintaining extreme sample environments at ORNL and a sophisticated polarized-beam triple-axis spectrometer at BNL. This equipment serves a variety of experimental needs and can be used for challenging experiments that broaden the field of neutron scattering. Although relatively modest in size, this program has been highly successful. It has produced some outstanding research, and it has also produced some very close interactions between neutron scattering scientists in Japan and the United States.

1. INTRODUCTION

The U.S.-Japan Cooperative Program on Neutron Scattering was negotiated under the Agreement between the Government of Japan and the Government of the United States of America on Cooperation in Research and Development in Science and Technology. This agreement was signed May 1, 1980, and the neutron scattering program was initiated in 1982. It involves two arrangements between the United States Department of Energy (DOE) and appropriate organizations of the Japanese government.

One of these arrangements is between DOE and the Science and Technology Agency of Japan (STA) and concerns research by scientists of the Japan Atomic Energy Research Institute (JAERI); this part of the program is managed by JAERI for the STA. The other arrangement is between DOE and the Japanese Ministry of Science, Education, and Culture (Monbusho); this part of the program is managed by the Institute for Solid State Physics (ISSP), University of Tokyo, for the Monbusho.

2. PROGRAM INFORMATION

The U.S.-Japan Cooperative Program on Neutron Scattering involves research collaboration by Japanese scientists with scientists at Oak Ridge National Laboratory (ORNL) and Brookhaven National Laboratory (BNL) and the construction of new neutron scattering equipment at both laboratories with funds provided by the Japanese government. The operating costs of the joint program, exclusive of neutrons, are shared equally by the two countries, with the United States providing neutrons in exchange for the new equipment provided by Japan. The annual assignment of Japanese scientists to ORNL and BNL corresponds to about two person years at each laboratory, and an equal number of U.S. scientists participate in the research. The Japanese assignments vary in length, and about fifteen scientists from Japan perform research in the United States annually under this program. The research program at BNL involves university scientists from Japan, and the research program at ORNL includes both university and JAERI scientists.

Two steering committees, an ISSP-DOE Steering Committee and a JAERI-DOE Steering Committee, are responsible for implementation of this cooperative program. These committees meet jointly once each year to review the research programs, to approve the research and budgets proposed for the next year, and to resolve any priority issues. Specific details of the programs are implemented by research committees at each laboratory that meet annually to review the research and to plan experiments and participants for the coming year. Changes in the planned research can be made during the year, but they must be approved by members of the research committee.

The main research collaboration is centered around the new equipment provided by Japan at the ORNL High Flux Isotope Reactor (HFIR) and at the BNL High Flux Beam Reactor (HFBR). This new equipment serves a variety of experimental needs and can be used for challenging experiments that broaden the field of neutron scattering. However, research investigations are not restricted to the new equipment, and other existing facilities are utilized when they are needed.

3. RESEARCH EQUIPMENT

3.1 Equipment provided for the ORNL-JAERI program

Under the ORNL-JAERI part of the program, the JAERI has provided a new type of wide-angle neutron diffractometer (WAND) at the HFIR [1,2]. Figure 1 is a drawing of this diffractometer, which uses a newly developed curved one-dimensional position-sensitive detector with an angular coverage of 130°. In order to reduce the detector background, a multiblade radial collimator with a fan shape is mounted in front of the detector inside the shield; this collimator is oscillated with constant velocity during measurements to smear out shadows of the blades.

Such detectors have previously been used mainly for powder measurements, but the application of such detectors in single-crystal diffractometry shows considerable promise. A very favorable technique for single-crystal experiments is to arrange the detector so that the whole detector arc lies on a specific layer of the diffraction pattern in reciprocal space; this is the geometry utilized in x-ray Weissenberg cameras. In the new instrument the crystal goniometer and the detector can be tilted together automatically to provide the proper geometry.

Time-resolved measurements are performed synchronously with the change of external sample conditions. One-shot measurements are made for phenomena that can not be repeated easily, and periodic measurements are employed in cases where the change is reversible according to a periodic external condition. Important auxiliary equipment, which has been provided by the JAERI for the WAND, includes a stress-modulation machine and a furnace for very rapid temperature changes [3]. The furnace permits increases in temperature between 200°C and 800°C at a rate of about 1000°C/min and decreases in temperature at a rate of about 700°C/min.

3.2 Equipment provided for the ORNL-ISSP Program

Under the ORNL-ISSP part of the program, the ISSP has provided equipment [4] for maintaining samples under extreme conditions, and this equipment can be used on nearly all neutron scattering facilities at the HFIR. This equipment includes an ultra-low temperature cryostat, high-pressure cells that can be used down to liquid helium temperatures, and two high-temperature furnaces for special applications.

The ultra-low temperature cryostat makes use of a He^3-He^4 dilution refrigerator, which produces temperatures down to about 50 mK for a powder specimen and to about 7 mK for a single crystal. The cryostat is equipped with a Helmholtz-type superconducting magnet, which provides a magnetic field up to 5 T with a homogeneity of 0.5% in a 2 cm^3 volume at the sample position. The high pressure cells that have been developed [5] are modeled after the design of McWhan and his associates [6]. Pressures are generated at room temperature using Fluorinert as the pressure-transmitting fluid, and pressure variations at cryogenic temperatures are estimated by measuring the lattice parameter of NaCl. Pressures up to 31 kilobars have been obtained at liquid helium temperatures. One high temperature furnace has been especially designed for in situ crystal growth and associated neutron scattering experiments at temperatures up to 1500°C. The other furnace allows uniaxial stress to be applied to the sample to study the effects of stress on crystallographic and magnetic properties.

3.3 Equipment provided for the BNL-ISSP Program

Under the BNL-ISSP part of the program, an advanced polarized neutron spectrometer has been provided by the ISSP at the High Flux Beam Reactor. The design [7,8] has been chosen to employ a triple-axis Tanzboden system with optional time-of-flight capability for advanced polarization modulation techniques.

The in-pile plug with 3-position steel rotary shutter-collimator assembly, monochromator housing drum, and the drum-supporting saddle are similar to those of other BNL triple-axis spectrometers except for minor changes and a larger beam size. In-pile collimators can be positioned automatically, and large filter assemblies, which can be cryogenically cooled, are located in the saddle shielding. The monochromator rotating drum provides a take-off angle from -2° to 108° ($2\theta_m$) and has a sufficiently large cavity to accommodate movable magnetic guides or multilayer polarizer systems. Either a pyrolytic graphite monochromator or a Heusler-alloy polarizing monochromator can be moved into position accurately by remote controls.

Sample and analyzer tables at the respective axes are on airpad assemblies to move on the Tanzboden floor. The $2\theta_m$ arm, which connects the monochromator drum to the sample table, is of sectional design to allow a large variation of this distance, and the sample-analyzer distance is also adjustable. Various angular rotations are driven by DC stepping motors and these orientations are measured by encoders in 0.01° increments.

The Tanzboden floor is made of low viscosity epoxy resin and is flat within a few hundredths of a centimeter, as required for proper functioning of the airpads. The entire spectrometer operation, including changing and monitoring certain sample conditions, is computer controlled, and a graphical display can be obtained after each measurement.

The spectrometer is now being modified to allow neutron scattering experiments by the neutron spectral modulation (NSM) technique. This technique combines spectral modulation of the incident neutron beam and correlation time-of-flight spectrometry. It is similar to the neutron spin echo technique in utilizing neutron spins to differentiate various incident neutron wavelengths; in NSM the neutron spin phase angles are converted to the incident neutron intensity modulation. It is anticipated that these modifications will be completed within a year.

4. RESEARCH INVESTIGATIONS

4.1 Research in the ORNL-JAERI Program

The ORNL-JAERI research program has concentrated on the utilization of the WAND. This instrument has been used very effectively for a variety of experiments.

The WAND has the capability of collecting diffraction patterns of polycrystalline samples very quickly, and time-dependent phenomena in such samples can be measured efficiently [9]. Typical of such investigations is a study of the ordering kinetics of Ni_3Mn [10]; this study also utilized the rapid-temperature-change furnace so that the process could be repeated quickly many times to obtain good counting statistics. Results show that as the temperature of Ni_3Mn is lowered abruptly from 600°C to 470°C, through the order-disorder transition at 510°C, the amount of order increases relatively slowly at first, then becomes faster, and finally saturates. It is believed that the slow growth is associated with the formation and coalescence of clusters, and the faster growth involves migration of domain walls.

The WAND is also very useful for studying single crystal samples, because a two-dimensional diffraction pattern over a wide range can be observed quickly by step scanning of the sample orientation. One of the most useful applications of wide-angle neutron diffractometry is in the field of magnetism. One-dimensional diffraction rods reflecting two-dimensional magnetic ordering, two-dimensional planes reflecting one-dimensional magnetic ordering, and magnetic diffuse scattering can be measured very effectively [11]. It is also possible to search efficiently for unknown satellite reflections caused by magnetic structures that are incommensurate with the chemical structure.

4.2 Research in the ORNL-ISSP Program

Various types of experiments have been performed in the ORNL-ISSP Program with the sample under extreme conditions. These experiments include investigations at milli-Kelvin temperatures of hyperfine-enhanced nuclear-spin ordering in singlet electronic ground state compounds [12]. The direct interaction between nuclei is extremely small and leads to cooperative nuclear spin ordering in the micro-Kelvin temperature region. However, in these compounds the hyperfine interaction admixes some electronic moment into the nuclear substates of the electronic ground state, producing an enhanced moment that can order at higher temperatures. The sensitivity of neutrons to the state of the nuclear-spin order arises from the spin dependence of the neutron-nucleus interaction. In $PrSn_3$, which orders antiferromagnetically at 8.6°K, additional Bragg peaks were observed below about 400 mK, which are due solely to the nuclear spin polarization of ^{141}Pr caused by the hyperfine field from the ordered electronic moments. The temperature dependence of the (3/2,0,0) peak intensity is shown in Figure 2. The solid curve, which is an excellent fit for the data, is a theoretical function that includes previously reported values for the hyperfine constant, nuclear spin, and saturated electronic moment. Magnetic phase transitions associated with the ordering of enhanced nuclear moments have also been observed in $PrCu_2$ and $HoVO_4$, at 58 mK and 4.5 mK, respectively. With $HoVO_4$ it was possible to reach a temperature of 2.7 mK through adiabatic demagnetization of the sample.

4.3 Research in the BNL-ISSP Program

Before the new triple-axis spectrometer was installed at the HFBR in 1985, many investigations were performed in the BNL-ISSP Program on other spectrometers that had been provided by the DOE. During the past two years nearly all of the research has utilized the new spectrometer. Most of the research at BNL has involved inelastic neutron scattering associated with structural phase transitions and with spin fluctuations in magnetic systems. Such studies have been made on a wide variety of magnetic materials that have included low-dimensional magnetic lattices, spin glasses, heavy-fermion systems, 3d metals and compounds, triangular-lattice antiferromagnets, and random magnetic systems.

Of particular interest and importance recently is a study [13] of the transverse fluctuations in an Ising spin glass, $Fe_{0.4}Mg_{0.6}Cl_2$. In this compound, which is an insulating material with a layered structure, there are competing ferromagnetic and antiferromagnetic interactions, and the easy magnetic axis is perpendicular to the layers. The spin-glass transition temperature T_{sg} has been determined to be 3.4°K, at which temperature the slow relaxations associated with the longitudinal spin component $S_{\parallel}$ (parallel to the easy axis) become frozen in random orientations. However, spin waves can be associated with the transverse component $S_{\perp}$, and Figure 3 gives the in-plane spin-wave dispersion at a temperature of 1.70°K. Although theoretical calculations have indicated that a system of spins ordered in random directions can have spin waves, this is the first observation of long-wavelength spin waves in a spin-glass system. Data for pure $FeCl_2$ at 5°K, which is well below the Néel temperature of 23.5°K, are shown in the figure for comparison. Both systems show a sharp energy gap at the center of the Brillouin zone (h=0), and by comparison with $FeCl_2$, it is believed that the energy gap in $Fe_{0.4}Mg_{0.6}Cl_2$ is due exclusively to single-ion anisotropy. The spin waves have been observed at temperatures up to about 2 T_{sg}, and the spin-spin correlation lengths both parallel and perpendicular to the easy axis have been determined. A simple explanation, involving the precession of spin clusters, has been proposed to explain these observations.

5. GENERAL COMMENTS

The U.S.-Japan Cooperative Program on Neutron Scattering has been in existence for over five years. Although relatively modest in size, it has been a highly successful program. Even though some cooperative research has been performed throughout all five years, much time was spent during the early years on the design, construction, installation, and testing of new equipment. Nevertheless, during calendar years 1984-86, about 50 papers associated with this program were published in scientific journals. Therefore, by almost any standards, the program has been very productive. Perhaps even more important, it has produced some very close interactions between scientists in Japan and those in the United States.

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REFERENCES

[1] Iizumi, M. et al., Progress Report on JAERI-ORNL Cooperative Neutron Scattering Research, April 1, 1984–March 31, 1985 JAERI-M85-112 (1985) 5.
[2] Morii, Y., Funahashi, S., and Child, H. R., Progress Report on JAERI-ORNL Cooperative Neutron Scattering Research, April 1, 1985–March 31, 1987 (1987) 3.
[3] Katano, S., Motohashi, H., and Iizumi, M., Rev. Sci. Instrum. 57 (1986) 1409.
[4] Kunitomi, N., Annual Report of the Japan-U.S. Cooperative Research Program on Neutron Scattering (ISSP-BNL/ORNL) JFY 1984 (1986) 9.
[5] Onodera, A. et al., Jap. J. Appl. Phys. 26 (1987) 152.
[6] McWhan, D. B. et al., Phys. Rev. B20 (1979) 4612.
[7] Ito, Y. and Shirane, G., Annual Report of the Japan-U.S. Cooperative Research Program on Neutron Scattering (ISSP-BNL/ORNL) JFY 1984 (1986) 3.
[8] Ito, Y. et al., Annual Report of the Japan-U.S. Cooperative Research Program on Neutron Scattering (ISSP-BNL/ORNL) JFY 1985 (1987) 3.
[9] Iizumi, M., Physica 136B (1986) 36.
[10] Katano, S. et al., Progress Report on JAERI-ORNL Cooperative Neutron Scattering Research, April 1, 1985–March 31, 1987 (1987) 14.
[11] Funahashi, S., Morii, Y., and Child, H. R., J. Appl. Phys. 61 (1987) 4114.
[12] Nicklow, R. M. et al., J. Appl. Phys. 57 (1985) 3784.
[13] Wong, Po-zen et al., Phys. Rev. Lett. 58 (1987) 1276.

FIGURE CAPTIONS

Figure 1. Schematic drawing of the Wide-Angle Neutron Diffractometer (WAND).

Figure 2. Temperature dependence of the peak intensity of the (3/2,0,0) diffraction peak from $PrSn_3$.

Figure 3. In-plane spin-wave dispersions for $Fe_{0.4}Mg_{0.6}Cl_2$ and $FeCl_2$.

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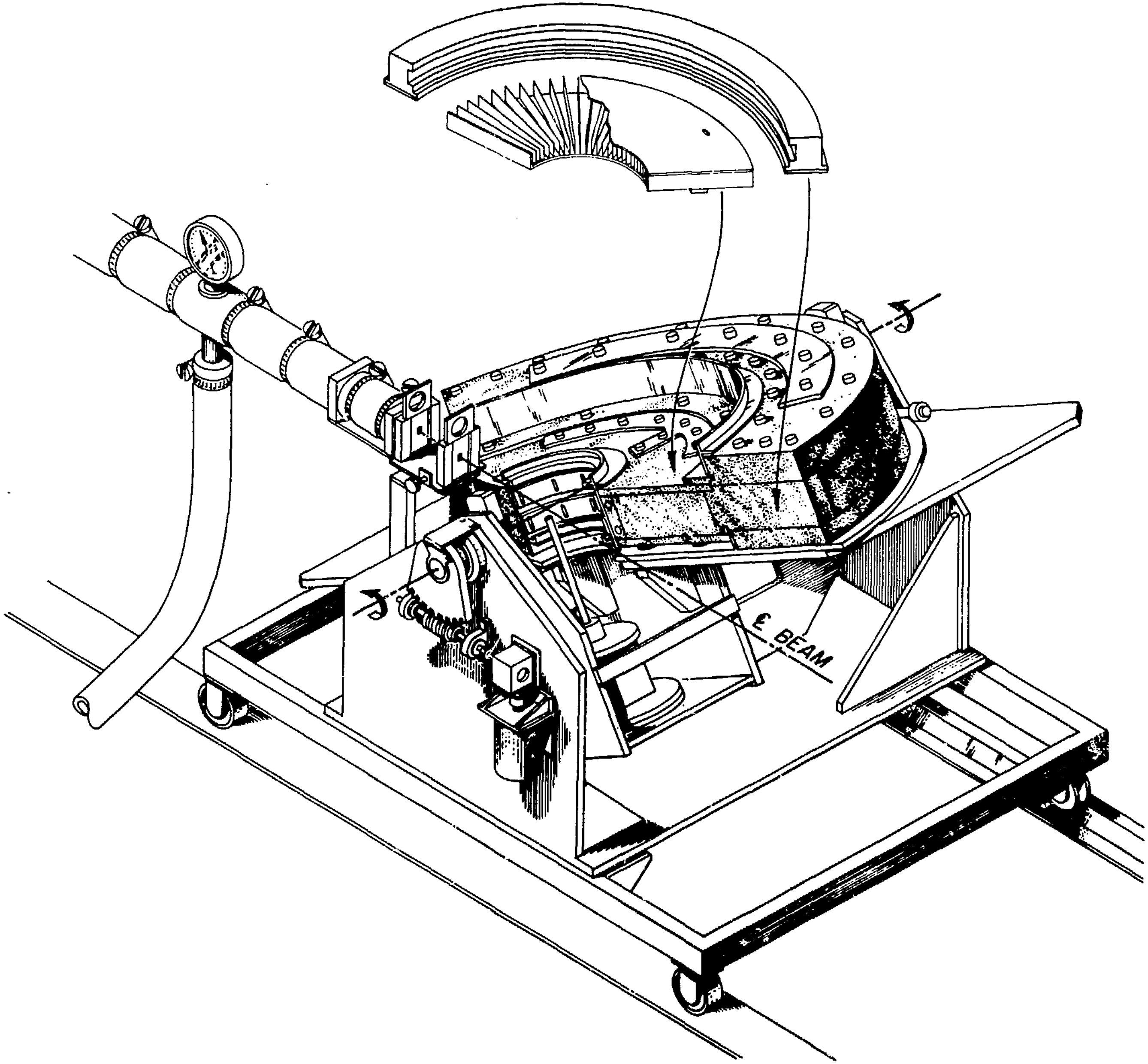

Figure 1. Schematic drawing of the Wide-Angle Neutron Diffractometer (WAND).

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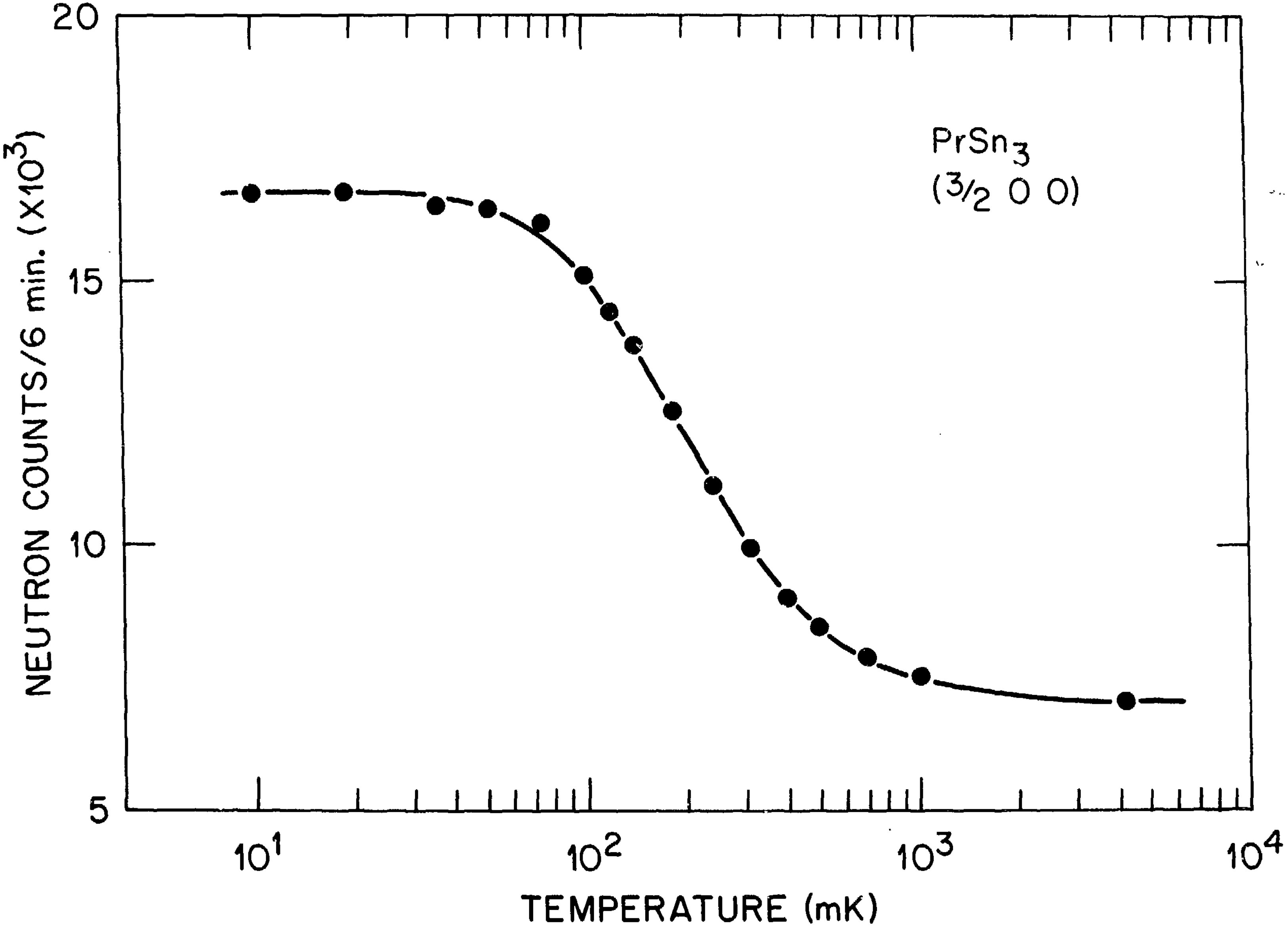

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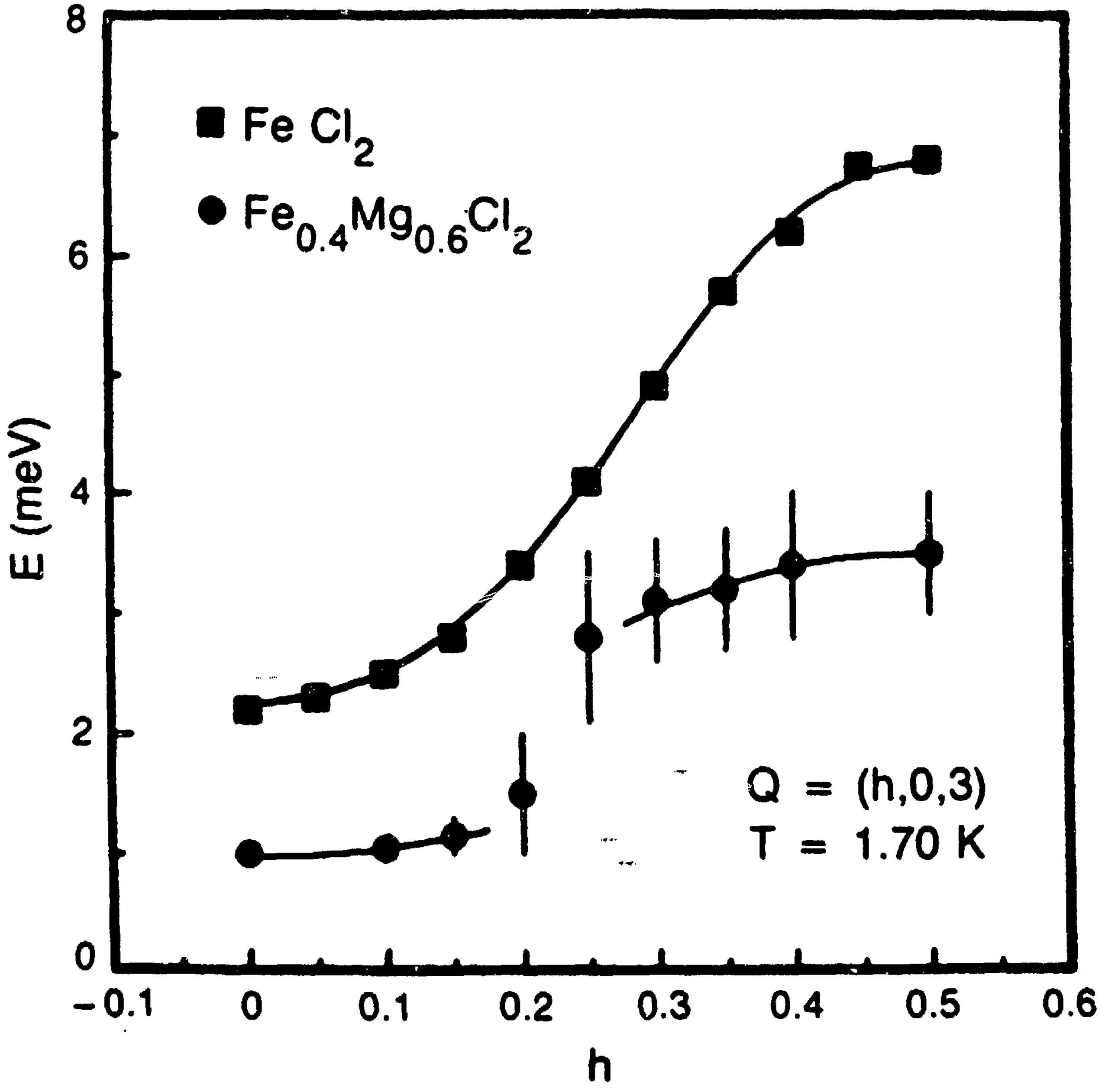

Figure 3. In-plane spin-wave dispersions for $Fe_{0.4}Mg_{0.6}Cl_2$ and $FeCl_2$.