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Calibration of Uniaxial Stress in the $KFeF_3$ Crystal Used in the Previous Mössbauer Measurement

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Abstract

The unknown stress in $KFeF_3$ crystal which was glued to an acrylic plastic plate and cooled below the Néel temperature T_N was calibrated by applying known pressure. The pressure partly cancelled the stress causing the increase of T_N. It was observed that the pressure of 1.7 ± 0.2 kbar decreased T_N by 3.7 ± 1.0 K. From this result it was estimated that in the stressed crystal, labeled "S-(111)-fixed", previously examined, the stress of about 2.5 kbar perpendicular to the glued plane caused the increase of T_N by 5 K. Pressure dependences of hyperfine field H_{hf} and $e^2qQ/2$ at 77 K were also obtained.

§ I. Introduction

In our previous work,[1),2),3)] in order to determine the direction of an antiferromagnetic easy axis, we used in Mössbauer measurements a single crystal of $KFeF_3$ consisting of a single domain which was produced by cooling below T_N under uniaxial stress. The stress was automatically caused at low temperature in the crystal glued to an acrylic plastic plate at room temperature, because of the difference between thermal expansion of the crystal and that of the plastics. Such stress caused effectively an elongation of the crystal along the direction perpendicular to the glued plane. In the stressed samples a considerable increase of the Néel temperature T_N was observed. A larger increase of T_N was observed for a larger thickness of the plastic plate relative to that of the crystal. The increase of T_N amounted indeed up to 8 degrees in the sample with a relative thickness of 30 (a crystal; 70 μm, a plastic plate; 2050 μm). The dependence of T_N on a uniaxial stress is considered to give an important information about exchange interactions. However, even the order of the uniaxial stress automatically caused in the glued crystal at low temperature has not been known and the relationship between the measured increase of T_N and the stress has not yet been established. We want to know, at least, the order of the stress caused in the glued crystal at low temperature.

The purpose of the present experiment is to calibrate this unknown stress. We applied known uniaxial pressure to the stressed crystal partly to cancel the stress causing the increase of T_N, and observed the change of T_N by Mössbauer spectroscopy.

§ II. Experimentals, results and discussions

A sample holder equipped with a pressure system was prepared for Mössbauer measurements at low temperatures. The holder must be small in size to be fitted to the dewar and must allow γ-rays to pass through. The holder consists of a pair of metallic plate, a crystal glued to an acrylic plastic plate, a thin acrylic plastic plate and four pairs of nuts and bolts made of brass. We applied pressure to the crystal by tightening the pair of metallic plates with the aid of bolts and nuts.

Details are shown in Fig. 1.

(a) A copper plate. It is 31 mm wide and 5 mm thick and is connected to the liquid N_2 bath through some thermal resistance. A heater wire is wound around the upper part of the copper plate and is used to change temperatures of the sample.

(b) A brass plate. It is 31×33 mm square and 4 mm thick. Each of (a) and (b) has a hole of 4 mm diameter at its center for transmission of γ-rays and four holes at the corners to get the bolts through.

(c) and (d) Four specially polished bolts (c) and nuts (d) of 8 mm dia-

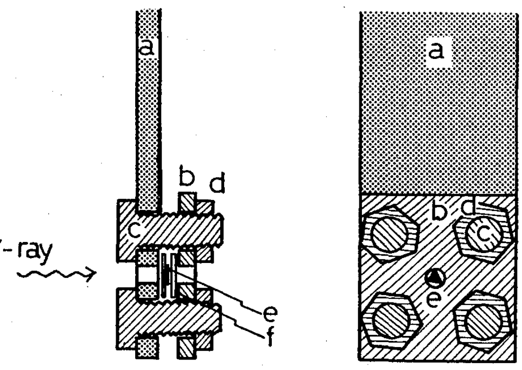

Fig. 1. A sample holder with a pressure system
(a) a copper plate.
(b) a brass plate.
(c) and (d) brass bolts and nuts.
(e) a crystal glued to an acrylic plastic plate.
(f) an acrylic plastic plate.
In the left figure, γ-rays pass through a crystal from the left side to the right side. In the right figure, γ-rays pass through a crystal perpendicularly to the paper.

meter made of brass. The contact planes between nuts and the brass plate and those between bolts and the copper plate are polished to decrease undesirable friction for good reproducibility.

(e) A (111) platelet of $KFeF_3$ crystal. It is glued to an acrylic plastic plate with epoxy resin.

(f) An acrylic plastic plate of 1 mm thickness. It is used to transmit pressure to the crystal.

The uniaxial pressure applied to the crystal was controlled by the strength of a torque required for tightening the metallic plates. For applying the pressure perpendicularly to the crystal plane, the special precautions were taken. The metallic plates were first tightened by wrenching the four bolts and nuts by hand and then by a wrench with a torque meter mentioned below, while we checked parallelism of the metallic plates frequently by measuring their gap at three fixed points.

The relationship between the torque and pressure (load) was calibrated as follows. The bolts were wrenched by a wrench with a torque meter

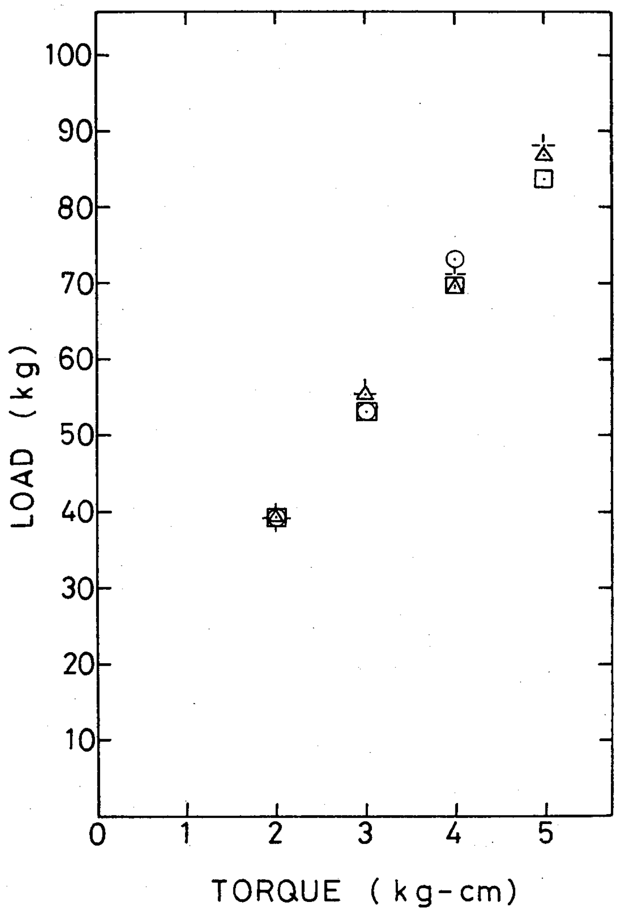

Fig. 2. The relationship between torque and load. Measurements were repeated four times. They were distinguished by square, triangle, circle and cross symboles.

of full scale of 40 kg-cm. We estimated load applied on a crystal by using a miniature load cell. (Produced by KYOWA Electronic Instruments Co., Ltd.) A load cell is a device to measure load using a strain gauge attached inside itself. A load cell chargeable up to 100 kg was used. The cell had a diameter of 20 mm and was not possible to be inserted into the sample holder (Fig. 1) instead of a crystal. The second holder 1.3 times large in linear dimensions except for the thickness compared with the sample holder was made in order to obtain the relationship between load and torque. The relationship between load and torque obtained by using the second holder is shown in Fig. 2. Measurements were repeated four times for torques of 2, 3, 4 and 5 kg-cm. Each measurement is shown by the symbol of square, triangle, circle or cross. We can see that load depends linearly on torque and that reproducibility of the relationship between load and torque is good. A torque of 5 kg-cm corresponds to a load of 88 kg as shown in Fig. 2. It is confirmed as follows that this relationship obtained for the second holder holds also for the sample holder used in Mössbauer measurements. The third holder 1.5 times large in linear dimensions except for the thickness compared with the second one was prepared. For the third holder we obtained the same relationship between torque and load as that shown in Fig. 2 within experimental errors using the same load cell. This fact is considered to show that the two metallic plates of the third holder did not bend by load of 88 kg (torque of 5 kg-cm) which was applied to the load cell. Because the two metallic plates of the sample holder are smaller than those of the second and the third holder, they are considered not to bend up to load of 88 kg, too. Consequently it was considered that the relationship shown in Fig. 2 holds also for the sample holder used in Mössbauer measurements. In order to decrease appreciably the stress causing the increase of T_N in the glued crystal used in this work we applied pressure corresponding to torque of up to 20 kg-cm. This value corresponds to about four times of the maximum value calibrated in the present work shown in Fig. 2. We extrapolated linearly the calibration curve four times in order to obtain the value of load applied to the crystal by 20 kg-cm torque. The value of the load of 350 kg estimated from this linear extrapolation is considered to be correct from the following reason. In the preliminary experiment, we used acrylic bolts and nuts. When a sample holder tightened at room temperatue with acrylic bolts and nuts was cooled down to 77 K, torque was measured to be twice of that applied at room temperature. On the other hand with brass bolts and nuts, torque at 77 K was measured to be unchanged. For acrylic bolts and nuts, the torque of 5.5 kg-cm applied at room temperature (11 kg-cm at 77 K) was estimated to correspond to the load of 350 kg at 77 K from the extrapolation of the calibration curve for acrylic bolts and nuts. For brass bolts and nuts the torque of 20 kg-cm applied at room temperature

(20 kg-cm at 77 K) was estimated to correspond to the load of 350 kg at 77 K from the extrapolation of the calibration curve in Fig. 2. We measured the hyperfine field for a stressed crystal $KFeF_3$ (mentioned later) at 77 K using Mössbauer spectroscopy in two cases. In one case pressure was applied to the crystal using acrylic bolts and nuts by torque of 5.5 kg-cm and in the other case pressure was applied to the same crystal using brass bolts and nuts by torque of 20 kg-cm at room temperature. The same value of the hyperfine field was obtained in both cases. This fact shows that the same load was applied to the crystal in both cases at 77 K. Consequently it is considered that the linear extrapolation mentioned above is reasonable, that is, the load depends linearly on torque up to four times of the value shown in Fig. 2.

We calibrated the stress by measuring the change of T_N of the stressed $KFeF_3$ crystal under the known pressure by Mössbauer measurements. In the present experiment we used a (111) platelet of $KFeF_3$ of 30 μm thick glued to an acrylic plate of 500 μm thick. The area of 22 ± 2 mm^2 sustained the applied load (including area of epoxy resin around the crystal). T_N of this crystal was raised by 6 ± 1 K compared with that of a free crystal. In the stressed sample on which a uniaxial pressure was applied by the method mentioned above, no considerable spread of T_N and broadening of the Mössbauer spectral lines were observed. This fact shows that the applied pressure is considered to be uniaxial and homogeneous in the crystal. T_N was determined by measuring the transition of Mössbauer spectra from antiferromagnetic pattern to paramagnetic one. T_N in the stressed crystal decreased as pressure increased as expected, and approached to T_N of a free crystal as shown in Fig. 3. We obtained the pressure shown in Fig. 3

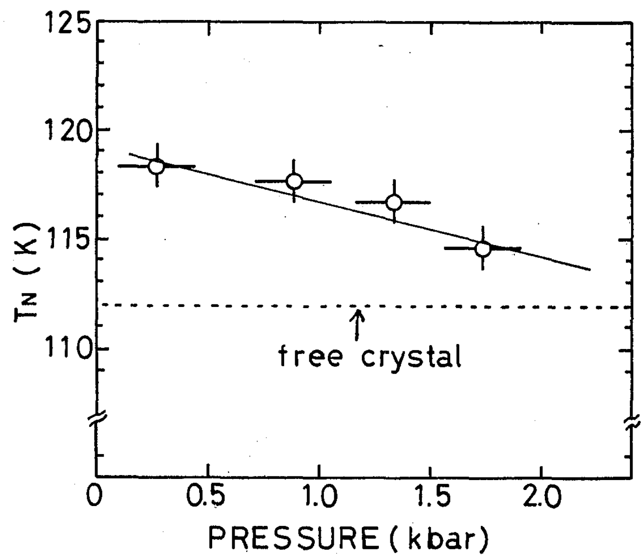

Fig. 3. The pressure dependence of T_N in the stressed crystal. A dotted line shows a value of T_N in a free crystal.

by dividing the load by the area of 22 mm^2. At the pressure of 1.7 ± 0.2 kbar, which is the highest in the present work, the decrease of T_N was 3.7 ± 0.1 K. In the crystal with T_N increased by 6 K used here, the stress of about 2.8 kbar was estimated to be applied perpendicular to the glued plane. To check the reproducibility of the process the crystal pressed to 1.7 kbar was released from pressure and pressed again to the same pressure. The same decrease of T_N was obtained. The spectra at 77 K were also observed under pressure. Both the values of the hyperfine field and $e^2qQ/2$ of the stressed crystal at 77 K, which differed from the value of the free crystal, approached the values of the free crystal respectively, as pressure increased.

Figures 4(a) and (b) show the pressure dependences of H_{hf} and $e^2qQ/2$ at 77 K, respectively. It is seen that both values of H_{hf} and $e^2qQ/2$ change linearly with pressure. This fact also shows that the linear relationship between torque and load can be extrapolated up to the torque of 20 kg-cm (1.7 kbar) as mentioned above. Both values of H_{hf} and $e^2qQ/2$ seem to

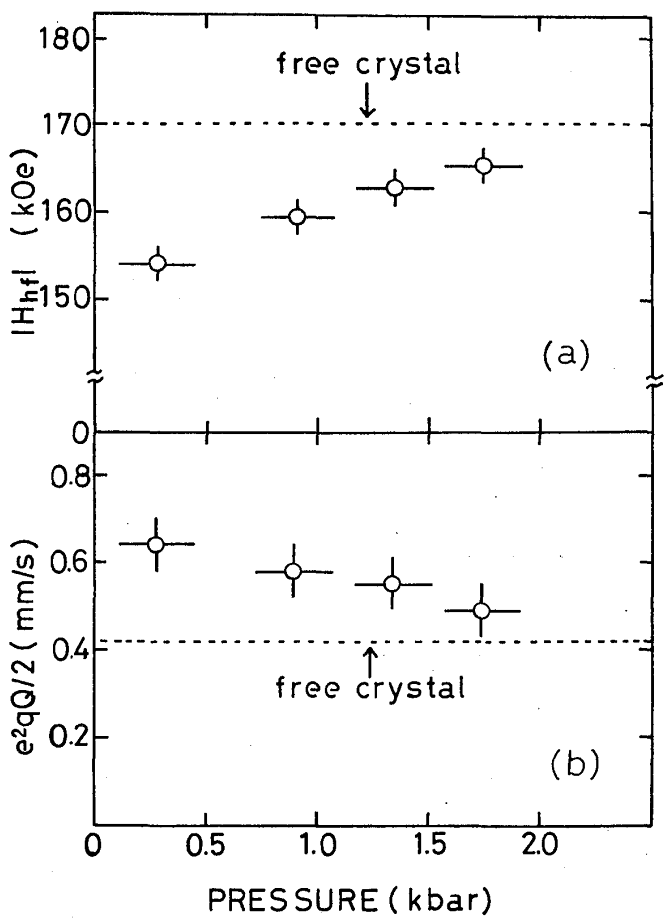

Fig. 4. The pressure dependence of the hyperfine field H_{hf} (a) and $e^2qQ/2$ (b) at 77 K in the stressed crystal. Dotted lines show the values of H_{hf} and $e^2qQ/2$ in a free crystal.

reach the values of the free crystal at about 2.3 kbar. But the value of T_N seems to reach the value of the free crystal at about 3 kbar if we extrapolate by a straight line through the data points within accuracy as shown in Fig. 3. The value of $dT_N/dp = -2.3 \pm 0.5$ K/kbar is obtained from a slope of the straight line. In an atiferromagnet MnO, Bloch et al. observed an increase of T_N at the rate of 3.8 ± 0.04 K/kbar[4] under a uniaxial stress (compression). Taking into consideration that $KFeF_3$ elongates along ⟨111⟩ direction but MnO contracts along ⟨111⟩ direction below T_N, the value of -2.3 ± 0.5 K/kbar obtained for $KFeF_3$ is considered to be reasonable including the sign.

In the stressed crystal with T_N increased by 6 K used in this experiment, an additional trigonal distortion is considered to add to a trigonal distortion appearing spontaneously below T_N. The temperature dependence of the lattice parameters α and a (lattice constant) in the stressed crystal is considered to be different from that in a free crystal.[5] Assuming that $T_N \propto J_{ex}(a, \alpha)$,* $J_{ex}(a, \alpha)$ of the stressed crystal is interpreted to be larger than that of a free crystal because T_N of the stressed crystal is larger than that of a free crystal.

The measurements could not be extended to higher pressure than 1.7 kbar because the limit of elasticity of the plastic plate (Fig. 1(f)) was reached and the results began to be irreproducible. The fact that three parameters (T_N, H_{hf} and $e^2qQ/2$) change with pressure linearly to 1.7 kbar confirmed that pressure was applied on the crystal in proportion to torque.

We showed that in the crystal with T_N increased by 6 K the stress of about 2.8 kbar was applied perpendicular to the glued plane. In the previous experiment[3] the crystal labeled "S-(111)-fixed" of 60 μm thickness which was guled to an acrylic plastic plate of 1 mm thickness was used to determine the orientation of the spin below T_N. We reported that the Néel temperature of the sample was increased by 5 K above that of a free crystal. From the result obtained in this work, the stress of about 2.5 kbar is considered to be applied in the crystal.

Acknowledgements

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* J_{ex} is the exchange constant.

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