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HAL Id: jpa-00218684
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Submitted on 1 Jan 1979

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MAGNETIC PROPERTIES OF THE PEROVSKITE COMPOUND RbFeF₃

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Abstract. - Mössbauer studies show that the spin axis in RbFeF₃ is along [111] in the three magnetic phases, which appear at $T_2 > T > T_1$, $T_2 > T > T_1$, and $T > T_1$, respectively. The weak ferromagnetism along [100] observed in the interval $T_2 > T > T_1$ is found to originate in a ferrimagnetic order of the two inequivalent Fe²⁺ sites. For the weak ferromagnetism along [110] observed in the interval $T > T_1$, the canting spin arrangement is suggested as its origin. The temperature variation of hyperfine magnetic fields is obtained from the ground state splittings.

1. Introduction. - The cubic perovskite RbFeF₃ has been shown to undergo successive magnetic and structural transformations at $T_N = 100.5$ K, $T_2 = 87$ K and $T_1 = 40$ K. It is antiferromagnetic in the interval $T > T > T_2$ and shows weak ferromagnetism in the two phases below $T_2$. Recently, we applied the Mössbauer spectroscopy on the stressed single crystal samples, and we found that the spin axis in the antiferromagnetic phase was along [111]. The spectra of powder samples observed below $T_2$ have not been analyzed unambiguously. In the interval $T_2 > T > T_1$, Wertheim et al. suggested that the weak ferromagnetism with an easy axis of [100] originates from a ferrimagnetic spin arrangement, since the Mössbauer spectra showed a superposition of two quite different hyperfine structures. On the other hand, Goodenough et al. suggested a canting spin arrangement as an origin of the weak ferromagnetism in the assumption that the Dzialoshinskii-Moriya interaction existed in this phase. The spin structures in both the phases below $T_2$, however, have not been determined experimentally.

In this paper we report the spin structure below $T_2$ and the temperature variation of hyperfine parameters.

2. Samples. - Stressed samples were prepared as follows. A thin slice of a single crystal with (111) surfaces and one with (100) surfaces glued at room temperature respectively to an acrylic plastic plate (a quartz plate) are expected to be compressed (stretched) in the glued plane with decreasing temperature and to experience a uniaxial elongation (compression) perpendicular to the plane. We labeled the stressed samples thus prepared as S-(111)-glued-P, S-(100)-glued-P and S-(100)-glued-Q. A powder sample and an unstressed single crystal sample with (100) surfaces (labeled as S-(100)-free) were also prepared.

3. Results and discussions. - 3.1. Spin structure. - The powder Mössbauer spectra are shown in figure 1, where (a), (b) and (c) are those characteristic of the three magnetic regions $T > T > T_2$, $T > T > T_1$ and $T < T_1$, respectively. In the interval $T_2 > T > T_1$, Wertheim et al. reported the same result, but the line assignment was not made by them. We successfully decomposed the complicated spectra into two different hyperfine structures due to two inequivalent Fe²⁺ sites. Wertheim et al. reported the same result, but the line assignment was not made by them. We successfully decomposed the complicated spectra into the two different hyperfine structures. The assignment indicated by I and II in figure 1 (b) was also supported by the observations under applied magnetic fields which were performed in order to investigate the origin of the weak ferromagnetism. The sample S-(100)-free was cooled down to 81 K in an applied field of 4 kOe along the [100] axis perpendicular to the sample plane, and its spectrum was taken with γ-rays parallel to the applied field. We obtained the spectrum in which the four lines indicated by circles in figure 1 (b), two of them belong to the site I and the others
to the site II, disappeared completely.

Fig. 1: Characteristic Mössbauer spectra of powder samples in the intervals (a) $T_1 > T > T_2$, (b) $T_2 > T > T_1$, and (c) $T < T_1$.

This fact shows that both the hyperfine field $H_{hf}$ and the spin in both the sites I and II are oriented in the direction of the applied field. Therefore, the weak ferromagnetism is definitely confirmed to originate in a ferrimagnetic order of the spin along a [$100$] in the site I and that in the site II. The Mössbauer spectra of S-(100)-glued-Q and S-(100)-glued-P were also observed with incident $\gamma$-rays perpendicular to the sample plane. For S-(100)-glued-Q we obtained the spectra lacking again the lines indicated by circles in figure 1 (b). On the other hand, in the spectrum of S-(100)-glued-P the intensity of those lines increased compared with that in the powder sample.

From the relative intensity of the absorption lines, we know that the spins are oriented in a direction [$100$] along which the lattice constant is the smallest (the crystal symmetry is believed to be orthorhombic in this phase but the detail has not been reported /5/).

In the interval $T < T_1$ the spectrum consists of six lines (see Fig.1(c)), and all Fe$^{2+}$ ions are regarded as equivalent. However, a possibility that the spectrum consists of two sextets almost overlapped has not completely been eliminated, because we notice a little but considerable broadening on some of the lines. Figure 2 shows the spectrum of S-(111)-glued-P observed with $\gamma$-rays perpendicular to the sample plane. The intensity of the spectrum is in good agreement with the calculated one (solid lines) on the assumption that the direction of $H_{hf}$ is parallel to that of $\gamma$-rays. Therefore, the spin axis is considered to be along [$111$] similarly to the antiferromagnetic region.

Fig. 2: Mössbauer spectrum obtained for S-(111)-glued-P with $\gamma$-rays perpendicular to the sample plane. Solid lines show the line intensity calculated on the assumption that the direction of $H_{hf}$ is parallel to that of $\gamma$-rays.

This fact suggests that the weak ferromagnetism along a [$110$] observed below $T_1$ /2/ originates in a canting spin arrangement. /1/

3.2. Hyperfine parameters. The temperature variation of $H_{hf}$ obtained from the ground state splittings is shown in figure 3.

Fig. 3: Temperature variation of $H_{hf}$ obtained from the ground state splittings. Although the hyperfine pattern changes drastically at the transition temperatures of $T_1$ and $T_2$, the value of $H_{hf}$ is not so much changed at $T_1$ and $T_2$ as seen in figure 3. Therefore, the drastic change of the patterns is attributed to the change in the value of $s^2qQ$ and in the relation between the directions of $H_{hf}$ and the principal axes of the EFG tensor. The unique set of the Mössbauer parameters has not been obtained for the spectra observed below $T_2$. The relative intensity of the spectra observed for single crystals is useful to determine the parameters. However, from the observations in the geometry that $\gamma$-rays are parallel to the direction of $H_{hf}$, one can not remove ambiguities in the values of $s^2qQ$, $\Theta_H$, $\phi_H$ and $\nu$ which are correlated to one another /5/. Further observations
are in progress using the geometries other than those mentioned above.

References


1) Note added in proof: Recently, we observed the intensity ratio of the spectrum at T<Tc in an applied magnetic field and it was determined for an unstressed sample that the spin axis was along [110] and the spontaneous magnetization originated from the canting of spins.