Laue diffraction of Mössbauer γ radiation from an Fe_3BO_6 crystal near the spin-reversal phase transition

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The energy spectra of the Laue diffraction of Mössbauer γ radiation in an Fe_3BO_6 crystal above and below the spin-reversal point T_{SR} are studied. Resonance peaks resulting from a combined hyperfine interaction have been detected in the (001) reflection spectrum. The effect of the spin-reversal and of the resonance-line interference on the diffracted-radiation spectra is discussed.

The compound Fe_3BO_6, an antiferromagnet with a weak ferromagnetism, is characterized by a complex structure of hyperfine magnetic and electric fields.[1–4] Its Néel point is $T_N = 508$ K and at the temperature $T_{SR} = 415$ K a spin-reversal phase transition occurs in this crystal. At $T < T_{SR}$ the antiferromagnetic axis lies along the [001] axis and a weak ferromagnetic moment lies along the [100] axis. At $T_{SR} < T < T_N$ the antiferromagnetic axis is oriented along the [100] axis and the weak ferromagnetic moment is oriented along the [001] axis.

The first experimental studies of Fe_3BO_6 using the diffraction of Mössbauer γ radiation[5–7] have made it possible to uniquely determine the particular features of the magnetic structure of the crystal and demonstrated that this method can be used effectively in the study of crystals with a complex structure of the hyperfine (magnetic and electric) fields. Furthermore, the interference of the scattering of Mössbauer radiation by iron nuclei, which occupy the crystallographically nonequivalent 4*c* and 8*d* positions, has been detected and thoroughly analyzed in these studies. These studies have also shown that this interference affects the energy spectra of the Bragg reflection.

In the present letter we report a study of the Laue diffraction of Mössbauer γ rays

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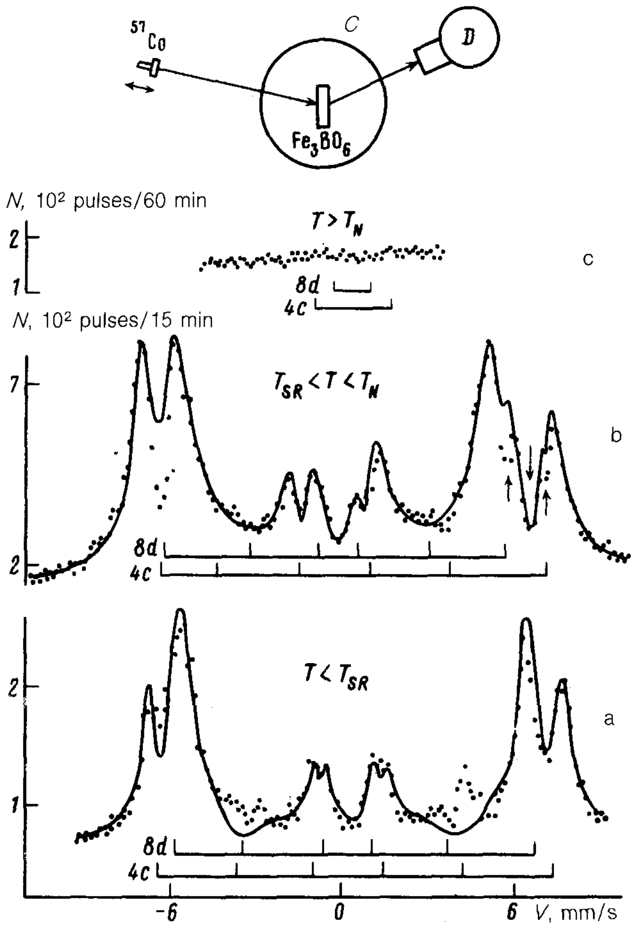

FIG. 1. The energy spectra of the Laue (001) reflection of Mössbauer γ radiation from an Fe_3BO_6 crystal. The spectra were measured at various temperatures: (a) 410 K, (b) 420 K, (c) 510 K. The solid curves are the results of numerical calculations carried out using an ideal-crystal model.

by an $^{57}Fe_3BO_6$ single crystal. To determine the effect of spin reversal on the intensity of the diffracted radiation, we carried out our measurements both below and above T_{SR}. We analyzed experimentally the spectra of the (001) reflection of Mössbauer 14.4-keV γ rays from an ^{57}Fe isotope at temperatures $T_1 = 410$ K and $T_2 = 420$ K.

The experimental arrangement is shown in the inset of Fig. 1. A beam of resonant γ rays from a $^{57}Co(Cr)$ source with an activity of ~ 300 μCi is incident on an $^{57}Fe_3BO_6$ crystal 15 μm thick. This crystal is placed into a temperature-controlled chamber C and aligned with the Laue symmetric (001) reflection. The beam divergence is 0.5° and the diffracted beam is detected by a semiconductor detector D. The chamber temperature is maintained within 0.01 K and the temperature gradient of the sample is no greater than 0.1 K. The results of the experimental studies and numerical calculations are shown in Fig. 1.

The most important structural feature of the experimental spectrum at $T = T_1 < T_{SR}$ (Fig. 1a) is the diffraction scattering of γ rays through the transitions with $\Delta M = 0$ (the second and fifth lines in the spectrum), where ΔM is the difference between the magnetic quantum numbers of the ground state and the excited state of the nucleus.[1)] An important point here is that at $T = T_2 > T_{SR}$ there is no evidence of a scattering through these transitions (Fig. 1b). We will show that diffraction scattering through the transitions with $\Delta M = 0$ cannot be explained exclusively in terms of the magnetic interaction or exclusively in terms of the quadrupole interaction, but is rather the result of a combined hyperfine interaction[8] in the Fe_3BO_6 crystal.

So far, only the magnetic structure of this compound has been determined.[2,6] Since no reliable data on the structure of the electric-field gradients have been obtained for Fe_3BO_6 (the results of the study of the structure in the Mössbauer transmission experiments[3] are at variance with the results of a symmetric analysis), the numerical calculations of the spectra were carried out with allowance of only the magnetic hyperfine interaction. The effect of the electric-field gradients on only the resonant line shift, which was determined from the transmission spectra, was taken into account in the calculations. In this approximation the (001) reflection is a purely nuclear magnetic reflection, for which there should be no scattering through the transitions with $\Delta M = 0$ (Ref. 9). The measured spectrum also corresponds to a purely nuclear reflection. A comparison of the theory with experiment shows that there is a good agreement among the spectra near the transitions with $\Delta M = \pm 1$ and that there is a discrepancy among them near the transitions with $\Delta M = 0$. Consequently, the scattering through the transitions with $\Delta M = 0$ cannot be accounted for exclusively in terms of the magnetic ordering.

The results of measurements at a temperature above the Néel point, i.e., under conditions when the magnetic order vanishes in the crystal but remains in the electric-field gradients have shown that there is no diffraction scattering in the (001) reflection (Fig. 1c); i.e., the scattering through the indicated transitions cannot be explained exclusively in terms of the quadrupole interaction.

We thus conclude that the resonant scattering of the Mössbauer γ rays through the transitions with $\Delta M = 0$ at $T < T_{SR}$ in the (001) reflection spectrum is due to a combined hyperfine interaction in the Fe_3BO_6 crystal. We have also observed the diffraction scattering through the transitions with $\Delta M = 0$ in the purely nuclear (005) and (007) reflections.

There is yet another curious feature in the spectra we have analyzed. This feature stems from the appearance in the Laue diffraction of the interference of the scattering of Mössbauer γ rays by ^{57}Fe nuclei situated in the nonequivalent positions (interference of the scattering by nuclei occupying the equivalent positions was discussed in Ref. 10 in the case of the $FeBO_3$ crystal). In the case of the (001) reflection, for example, we see a constructive interference[7] which leads to a broadening of the resonant lines and to a splitting of individual lines in the spectrum which is seen most clearly in the case of the outermost lines. The resonant-line splitting may have a peculiar "fine structure" if the energy spacing between the resonances corresponding to the same transition of the ^{57}Fe nuclei in the $4c$ and $8d$ positions is reasonably large. In particular, the sixth line in the spectrum in Fig. 1b, for which the spacing between the resonances of the iron nuclei in the $4c$ and $8d$ positions is $\approx 12\Gamma$, where Γ is the natural Mössbauer line width, exhibits three discrete minima indicated by the arrows. The two outer minima are attributable to the resonant absorption of the γ rays in the crystal and their positions coincide with the positions of the corresponding resonances for the iron nuclei in the $4c$ and $8d$ positions. The central minimum, which is caused by the interference in the scattering of γ rays, is the deepest minimum for the given thickness of the crystal.

We note in conclusion that we have observed for the first time a diffraction scattering of Mössbauer γ rays which results from a combined hyperfine interaction. The presence of this interaction at $T < T_{SR}$ and its absence at $T > T_{SR}$ is evidence that the scattering intensity depends strongly on the mutual orientation of the magnetic and electric fields in the crystal. We also note that the use of Mössbauer radiation diffraction in the Laue geometry to study phase transitions makes it possible to analyze the phase transition in the bulk of the sample. This approach is also less sensitive to the surface effects than the use of the Bragg geometry in similar studies.[11]

[1)]We are using here the classification of the transitions corresponding to the magnetic hyperfine interaction.

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