Purely nuclear diffraction of Mössbauer radiation in the critical region near the Néel point

G. V. Smirnov, M. V. Zelepukhin, and W. van Burk[1)]

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The Mössbauer spectra of the purely nuclear diffraction of resonant γ radiation of ^{57}Fe in the crystal $^{57}FeBO_3$ have been studied near the phase transition of the crystal from an antiferromagnetic state to a paramagnetic state. The spectra are shown to be sensitive to a nucleation of antiferromagnetic ordering of the atomic moments in the critical region of the transition.

The critical dynamic phenomena which occur in magnetic materials during second-order phase transitions have recently attracted considerably greater interest, primarily because of the theoretical progress, which has substantially improved our understanding of the critical dynamics of magnetic materials.[1,2] Another contributing factor has been the further development of experimental methods.[3–8]

In the present letter we propose a methodologically new approach to the study of critical magnetic phenomena in crystals. This approach is based on the phenomenon of purely nuclear diffraction of Mössbauer γ rays. This type of diffraction has an elevated sensitivity to the presence of a structural disorder of the fields in the crystal. Let us examine this question in more detail.

A decisive role is played in the formation of the diffraction pattern of radiation in a crystal by the phase relations among the waves which are scattered by the atomic sites of the crystal lattice. The wave phase relations are determined as geometric factors which depend on the atomic structure of the crystal and the nature of the scattering process. In the case of Mössbauer γ rays, the scattering by an atom is of a complex nature, consisting of the sum of two coherent processes: a resonant scattering by the nuclei of the Mössbauer isotope and a potential scattering by electrons. The interference of these two processes, each subject to its own phase relations, gives rise to a rather complicated diffraction pattern of the radiation in the crystal.

A feature of resonant nuclear scattering which is of importance to this question is the strong dependence of the phase of the wave scattered by a nucleus on the orientation of the nuclear spin with respect to the wave vectors. The spin orientation is related to the direction of the magnetic moment of the atom by virtue of the magnetic hyperfine interaction. For the electron scattering of the Mössbauer γ rays, there is no corresponding dependence. By virtue of these circumstances, nuclear resonant scattering—and it alone—is sensitive to the presence of a structural ordering of the magnetic fields in a crystal. The cases in which the atomic structure of the crystal does not coincide with its magnetic structure, intensity peaks are produced in the diffraction pattern exclusively as a result of the diffraction of the radiation by the nuclear lattice of the crystal. The angular positions and intensities of these purely nuclear reflections convey the symmetry of the ordering of the magnetic fields in the crystal. These purely nuclear reflections are of interest for research on phase transitions. The diffraction of Mössbauer γ rays by the nuclear lattice of a crystal was first observed in the antiferromagnetic crystal hematite.[9]

The effectiveness of this method in studies of the magnetic critical state of an antiferromagnet is based on the fact that the very existence of Mössbauer spectra in the case of purely nuclear diffraction results from an antiferromagnetic ordering of the spins in the crystal lattice. Near the Néel point, as the paramagnetic region is approached, the diffraction signal is quenched, providing direct evidence of a disruption of the antiferromagnetism in the crystal. This behavior of the diffraction spectrum is qualitatively different from the behavior of the spectra in conventional transmission Mössbauer experiments, where the presence of a residual magnetism is expressed only as a slight broadening of the resonant lines. We would therefore expect that purely nuclear diffraction would make it possible to carry out studies in the immediate vicinity of T_N and that the use of this new method to study phase transitions would provide additional information on the nature of the magnetic critical state of a crystal.

We have carried out a first experiment in this direction with the antiferromagnetic crystal $FeBO_3$. The Néel point in this crystal is only 50° above room temperature. The sample used for the measurements was grown at the Physics Institute of the Czechoslovak Academy of Sciences[10] from a material enriched to 95% in the Mössbauer isotope ^{57}Fe. The sample is a single-crystal plate with linear dimensions of $8\times6\times0.1$ mm. The sample is placed in a chamber for heating; the chamber is mounted on a goniometer. The temperature at the sample can be varied from 300 K to 500 K and regulated within 0.01 K; the temperature variation over the sample does not exceed 0.1 K. The design of the chamber allows the imposition of a magnetic field of up to several hundred gauss on the sample. We study the temperature dependence of the Mössbauer spectra of the purely nuclear diffractive scattering of γ rays from the (111) system of planes of iron borate in the third order of reflection. These planes run parallel to the surface of the crystal.

Figure 1 shows spectra measured at various temperatures near T_N in a 100-G magnetic field. As the temperature is raised, the spectrum obviously collapses, and the diffraction pattern simultaneously disappears. The experimental data at temperatures $T<T_N$ are described well by the theory with the value $T_N = 348.35$ K and with the static critical index $\beta = 0.37$, in agreement with the values found in Ref. 11.

The results of the measurements above T_N are of greatest interest. Here, unusual Mössbauer spectra are obtained in the presence of a field. These spectra, of a purely magnetic nature, consist of only a single line. Its total width is $\Gamma_s + \Gamma_n$, where Γ_s is the source linewidth, while Γ_n is the natural width of the nuclear level. In other words, in the crystal the width of the resonance under these conditions is approximately equal to the natural width of the nuclear level. The observation of a diffraction by the nuclear lattice of the crystal above T_N, by virtue of these distinctive features of this phenomenon, can be explained only on the basis of an antiferromagnetic ordering of the spins in the crystal. This type of ordering should in fact occur by virtue of the Dzyaloshinskiĭ interaction. This so-called effect of an antiferromagnetism induced by a magnetic field was predicted in Ref. 12 and verified experimentally in Refs. 13 and 14. The average magnetic field at the nucleus in this case is so weak that the magnetic

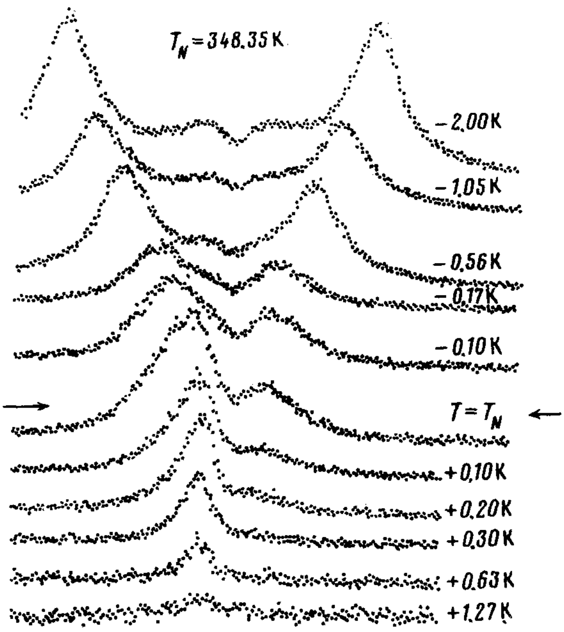

FIG. 1. Mössbauer spectra of purely nuclear diffraction of resonant γ radiation of ^{57}Fe in a $^{57}FeBO_3$ single crystal; the (333) Bragg reflection. An external magnetic field of about 100 G is applied to the crystal. The temperature is varied near the transition from the antiferromagnetic state to the paramagnetic state.

hyperfine splitting of the nuclear levels is hidden within the natural width of the level of the first excited state of the nucleus.

The restoration of antiferromagnetic order by a field in the parametric temperature region is observed in the present experiments (Fig. 2) as the external magnetic

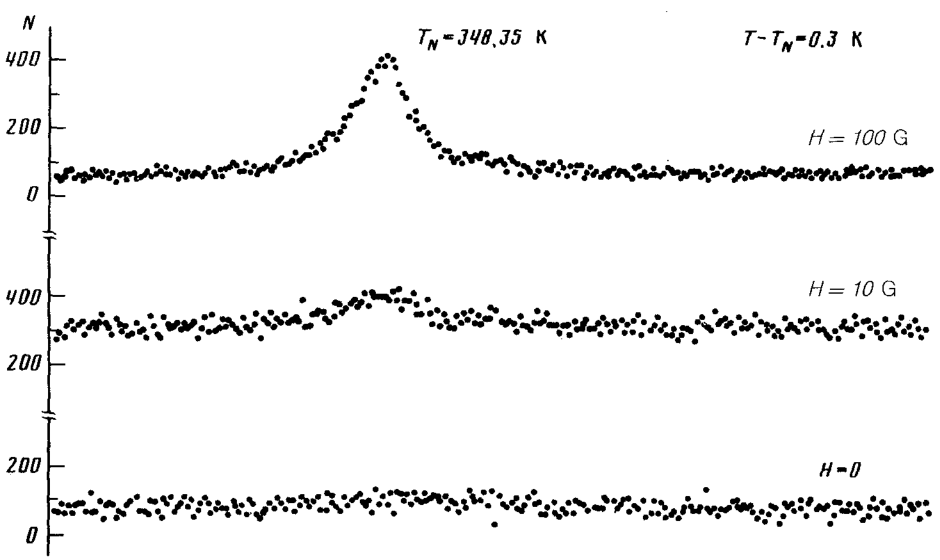

FIG. 2. Induction of spectra of purely nuclear diffraction of resonant γ rays by applying a magnetic field to a crystal.

field is lowered all the way to 10 G! It would be exceedingly difficult to observe effects of this sort in a transmission experiment, since the corresponding broadening of the resonant line would be no more than 0.1%. This circumstance is a convincing demonstration of the sensitivity of this new method to the nucleation of magnetic order in a crystal in the critical temperature region.

There is the interesting possibility of using this result to filter Mössbauer synchrotron radiation. The method of purely nuclear diffraction is most effective in solving problems of distinguishing a narrow band of resonant radiation from the continuous synchrotron spectrum.[15] However, it has heretofore been assumed that this method is capable of singling out only a complex spectrum consisting of several broadened lines. A spectrum of that sort was first found in Ref. 16. Our studies show that the use of nuclear diffraction at temperatures above T_N makes it possible to develop a synchrotron-radiation Mössbauer source which emits a single line with a width close to the natural width of the nuclear level.

[1)]Technical University, Munich, FRG.

[1]P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. **49**, 435 (1977).
[2]G. F. Mazenko and O. T. Valls, Phys. Rev. B **24**, 1419 (1981).
[3]C. Hohenemser, R. M. Suter, L. Chow, M. A. Kobeissi, R. Dunlap, and A. M. Gottlieb, Hyperfine Interaction **10**, 887 (1981).
[4]M. A. Kobeissi, Phys. Rev. B **24**, 2380 (1981).
[5]C. Hohenemser, L. Chow, and R. M. Suter, Phys. Rev. B **26**, 5056 (1982).
[6]F. Mezei, Phys. Rev. Lett. **490**, 1096 (1982).
[7]H. Keller and I. M. Savid, Phys. Rev. B **28**, 2638 (1983).
[8]J. Slivka, H. Keller, W. Kündig, and B. M. Wanklyn, Phys. Rev. B **30**, 3649 (1984).
[9]G. V. Smirnov, V. V. Sklyarevskiĭ, R. A. Voskanyan, and A. N. Artem'ev, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 123 (1969) [JETP Lett. **9**, 70 (1969)].
[10]M. Kotrbova, S. Kadeskova, N. Novak, J. Bradler, G. V. Smirnov, and Yu. V. Shvydko, J. Cryst. Growth **71**, 607 (1985).
[11]M. Eibschütz *et al.*, J. Appl. Phys. **41**, 1276 (1970).
[12]A. S. Borovik-Romanov and V. I. Ozhogin, Zh. Eksp. Teor. Fiz. **43**, 2042 (1960) [Sov. Phys. JETP **16**, 1443 (1960)].
[13]L. A. Prozorova, Zh. Eksp. Teor. Fiz. **57**, 1967 (1969) [Sov. Phys. JETP **30**, 1065 (1970)].
[14]S. S. Yakimov, V. I. Ozhogin, V. Ya. Gamlitskii, V. M. Cherepanov, and S. D. Pudkov, Phys. Lett. **39** A, 421 (1972).
[15]A. I. Checkhin, N. V. Andronova, M. V. Zelepukhin, A. N. Artem'ev, and E. P. Stepanov, Pis'ma Zh. Eksp. Teor. Fiz. **37**, 531 (1983) [JETP Lett. **37**, 633 (1983)].
[16]E. Gerdau, R. Rüffer, H. Winkler, W. Tolksdorf, C. P. Klages, and J. P. Hannon, Phys. Rev. Lett. **54**, 835 (1985).

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