

contribution of valence electrons to the stopping is always significant.

The theory presented in this work for stopping in an inhomogeneous electron gas can be applied also to the calculation of the total energy loss of channeled relativistic particles, where the stopping is determined by the effect of the density of the medium.

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## Features of the suppression effect under conditions of hyperfine quadrupole splitting

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We report the first experimental investigations of the effect of suppression of a nuclear reaction under conditions of hyperfine quadrupole splitting. The investigations were performed on a perfect iron borate crystal enriched with the resonant isotope  $Fe^{57}$ . A strong effect of the interference of different nuclear transitions on the form of the Mössbauer spectrum of the gamma-quantum Laue diffraction is observed.

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In our preceding articles<sup>1</sup> we reported investigations of the suppression effect (SE) of a nuclear reaction in an  $Fe^{57}BO_3$  crystal for pure nuclear magnetic diffraction of the Mössbauer rays. Besides the advantages of iron borate for the study of the SE described in Ref. 1, there are also other advantages that make it possible to expand the research on the effect, particularly to include the case of quadrupole splitting of the nuclear energy levels.

As was previously observed,<sup>2</sup> the iron borate crystal

has a large number of reflections in which complete or almost complete extinction of electron diffraction is obtained as a result of mutual cancellation of waves scattered by atoms of different species, iron on the one hand, and boron and oxygen on the other. For example, complete compensation is reached in the previously investigated<sup>2</sup> (222) reflection, where pure nuclear diffraction was observed for the first time ever in scattering of quanta on nuclear transmissions with  $\Delta m = 0$ .

Another advantage of the considered crystals is the

proximity of the point of the phase-transitions from the antiferromagnetic into the ferromagnetic state (the Néel point) to room temperature,  $T_N = 348$  K. Above  $T_N$  there remains in the iron-borate crystal only the quadrupole splitting of the nuclear energy levels with  $\Delta E = 3.9 \Gamma_n$  ( $\Gamma_n$  is the natural width of the nuclear level). On going through the Néel point, the pure nuclear diffraction of the quanta, which is connected with the antiferromagnetic ordering used in the preceding study,<sup>1</sup> vanishes. At the same time, the pure nuclear diffraction due to the extinction mechanism considered above still remains.

Thus, conditions are produced in the  $\text{Fe}^{57}\text{BO}_3$  crystal for the observation of pure nuclear diffraction in quadrupole splitting of nuclear levels, and for the investigation under these conditions of the distinguishing features predicted for the SE by the Kagan-Afanas'ev (KA) dynamic theory.<sup>3</sup>

It is known that the quadrupole interaction leads to a hyperfine splitting of the level of the  $\text{Fe}^{57}$  nucleus in the excited state into two sublevels, each of which is doubly degenerate. In the Mössbauer spectrum there are observed in this case two resonant lines. One line combines four nuclear transitions:  $\frac{1}{2} \rightarrow \pm \frac{1}{2}$  and  $-\frac{1}{2} \rightarrow \pm \frac{1}{2}$ , while the other combines two transitions,  $\frac{1}{2} \rightarrow \frac{3}{2}$ ,  $-\frac{1}{2} \rightarrow -\frac{3}{2}$ . According to the general theorem of the KA dynamic theory, when two transitions are combined in one resonance line, the SE should be observed simultaneously for two orthopolarized components of the gamma beam.<sup>3,4</sup> This circumstance, as well as the expected unusual manifestation of interference of nuclear transitions in this case, is the reason for the interest in it. For a long time it was impossible to observe the SE in quadrupole splitting, owing to the lack of suitable objects for the investigation. In the present paper we report the first experimental investigations of the features of SE under conditions of hyperfine quadrupole splitting, performed on iron-borate crystals.

### OBTAINING QUADRUPOLE PURE NUCLEAR DIFFRACTION BY AN $\text{Fe}^{57}\text{BO}_3$ CRYSTAL

The measurements were made with a Mössbauer diffraction spectrometer. The experimental setup is shown in Fig. 1a. The gamma beam coming directly from the source  $S$  (the same as in the preceding paper<sup>1</sup>) was restricted with collimators. Its divergence in the scattering plane was  $1^\circ$ . The scattered radiation was registered with a detecting system  $D$ .<sup>1</sup> The heating cell of the GPTV-1500 temperature chamber was modified to heat the crystal and was mounted on a goniometric head GP-3. With this combination we were able to adjust the crystal in the heated state, performing all the necessary adjusting displacements and rotations. The crystal was mounted on a porcelain post of 1.5 mm diameter with the aid of dental cement. The heating was directly in air to  $t = 80^\circ \text{C}$ . The temperature was monitored, accurate to  $1^\circ \text{C}$ , with a chromel-alumel thermocouple and an amplifier I37 with an automatic plotter N391. To introduce the gamma beam into the heating volume and to extract it we used a mica window  $40 \mu\text{m}$  thick.

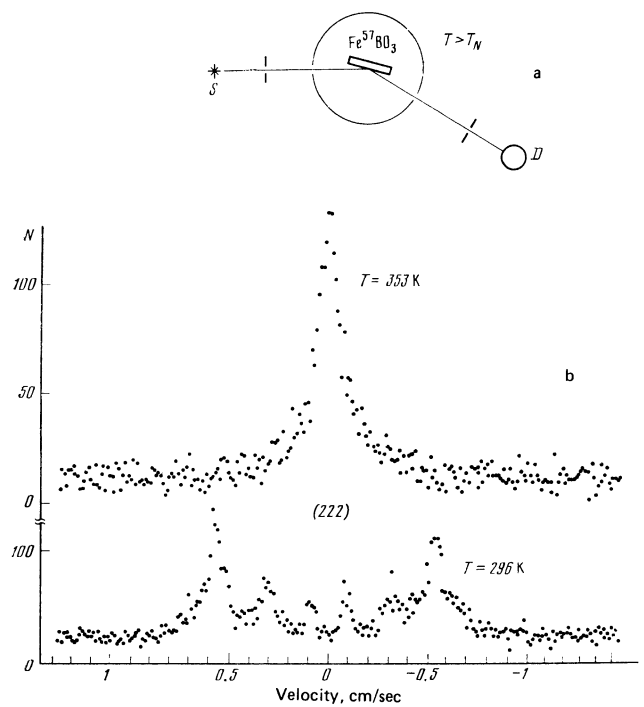


FIG. 1. Experimental setup and Mössbauer spectra of Bragg diffraction (222).

Figure 2 shows an x-ray diffraction topogram of the sample. It is seen that its central part has an extensive section in which the crystal lattice has a high degree of perfection. The rocking curves in this region, measured with  $\text{K}_{\alpha 1}\text{Mo}$  x-rays in a two-crystal combination  $\text{SiO}_2(20\bar{2}2)_{\text{Bragg}} - \text{Fe}^{57}\text{BO}_3(110)_{\text{Laue}}$ , had a width  $\sim 8''$ , which turned out to be not much worse than that expected for perfect crystals.

The transition beyond the Néel point in the investigated crystal was verified in the measurements of the Mössbauer spectrum of pure nuclear Bragg diffraction (222). Figure 1b compares the spectra obtained at two crystal temperatures, below and above the Néel point.

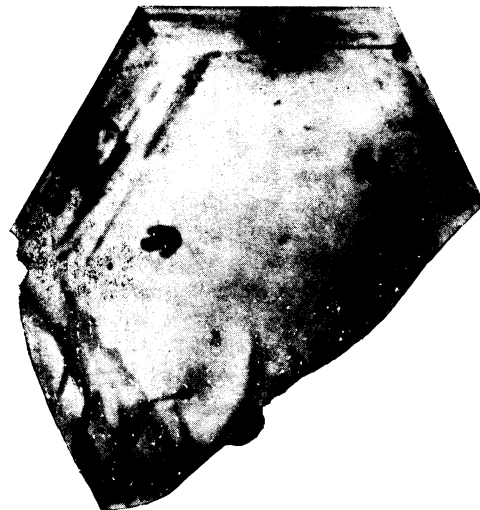


FIG. 2. X-ray diffraction topogram of  $\text{Fe}^{57}\text{BO}_3$  crystal (magnification  $30\times$ ).

The collapse of the six-line spectrum upon heating proves that the crystal becomes nonmagnetic. As to the quadrupole splitting, under conditions of Bragg diffraction it turned out to be unresolved because of the line broadening.<sup>5</sup> Its existence is indicated only by a certain symmetry in the contour of the maximum in the upper spectrum of Fig. 1b. We note that our measurements have revealed for the first time a magnetic phase transition under conditions of pure nuclear diffraction of Mössbauer beams.

### MEASUREMENTS OF LAUE DIFFRACTION

The isotopically enriched Fe<sup>57</sup>BO<sub>3</sub> crystals grown by the previously described method<sup>1</sup> were produced in the form of plates with the basal plane (111) on the surface. With this faceting of the samples, the best reflection from the point of view of separating nonmagnetic nuclear diffraction in the Laue geometry was (330). The (330) planes are inclined to the surface of the crystal plane at an angle  $\varphi = 61^\circ 4'$ , and the Bragg angle for reflection from these planes is  $\theta = 21^\circ 37'$  (Fig. 3). The amplitude of the coherent electron scattering for this reflection was 1.5–6% of the nuclear resonant amplitude, depending on the transition. The conditions were thus close to pure nuclear diffraction.

We used a scattering geometry with an asymmetry factor  $\beta = \cos \star \mathbf{k}_0 \cdot \mathbf{n} / \cos \star \mathbf{k}_1 \cdot \mathbf{n} = 0.64$ , which is more favorable, with respect to intensity, for a Laue beam in the  $\mathbf{k}_1$  direction. The electric field gradient (EFG) axis was directed along the normal  $\mathbf{n}$  to the surface of the crystal and was in the scattering plane. The resonance absorption factors  $(\mu t)_{res}$ , for ordinary passage of gamma quanta through the crystal near the  $\mathbf{k}_0$  direction, had the values listed in the table (the crystal was 75  $\mu\text{m}$  thick). It is seen that the crystal constitutes a strong resonant absorber for both polarizations. For an unpolarized gamma beam, both resonances turned out to be approximately equivalent with respect to absorptivity.

The angular crystal positions corresponding to the (330) reflection was first determined approximately with the aid of neighboring strong reflections (110) and (220), after which it was refined when the angle curve (330) was plotted. The crystal was then fixed at the maximum of reflection and the Mössbauer diffraction spectra were measured, first covering a large frequen-

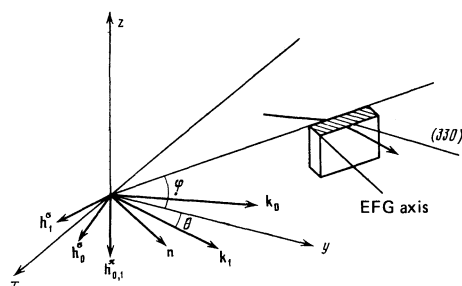


FIG. 3. Geometry of the vectors in the scattering scheme:  $\mathbf{n}$ —inward normal to the crystal surface;  $\varphi$ —angle between the entrance surface and the reflecting plane;  $\theta$ —Bragg angle;  $\mathbf{k}_0$  and  $\mathbf{k}_1$ —wave vectors of the incident and scattered waves;  $\mathbf{h}_{0,1}^r$  and  $\mathbf{h}_{0,1}^s$ —polarization vectors.

TABLE I.

Polarization	Transitions	
	$\pm 1/2 \rightarrow \pm 1/2$	$\pm 1/2 \rightarrow \pm 3/2$
$\pi$	100	300
$\sigma$	275	122

cy interval, so as to see the picture as a whole (Fig. 4), and then in a more detailed scale on the section  $v_1 - v_2$ , to study the variation of the intensity of the diffraction near the resonances (Fig. 5a). To determine the position of the resonance, we measured, at the same temperature, the usual Mössbauer spectrum in a thin resonant FeBO<sub>3</sub> absorber (Fig. 5b). The dashed lines show the resonant frequencies.

It is seen from the overall picture (Fig. 4) that the spectrum extends far beyond the limits of the resonance region. The resonances are separated by an intensity dip, and the maximum intensity is reached at distances  $11.5\Gamma_n$  and  $6.5\Gamma_n$ , respectively, on the right and left of the nearest resonances, i.e., where the crystal acquires already an effectively intermediate thickness ( $\mu t \approx 1$ ).

The most remarkable feature is the strong tilt of the spectrum of the transmission of the quanta through the crystal in favor of the region that covers the resonance  $\pm 1/2 \rightarrow \pm 3/2$ , even though, as already noted above, both resonances were approximately equivalent in absorptivity. The spectrum of Fig. 5a shows the difference in the intensities at the exact resonances. Another interesting result is that the minimum of the intensity occurs precisely in the interval between the resonant frequencies.

### ANALYSIS OF RESULTS

The case of Laue diffraction of resonant radiation in quadrupole hyperfine splitting was analyzed in detail theoretically by Afanas'ef and Kagan.<sup>3</sup> On the basis of the results of the KA dynamic theory, we now obtain for the coefficients of the dynamic equations in the considered case (Fig. 3) the following expressions (inasmuch as the electron scattering and absorption play a minor role in the formation of the superposition states, it is

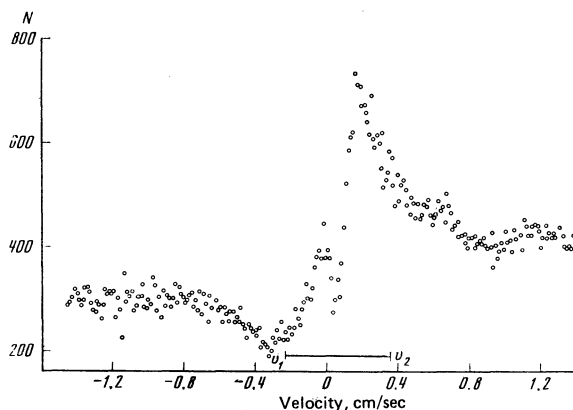


FIG. 4. General view of the Mössbauer spectrum of the (330) Laue diffraction.

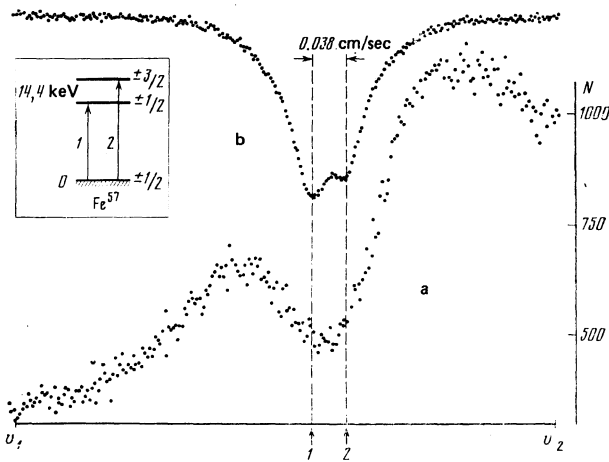


FIG. 5. a) Central part of the spectrum shown in Fig. 4; b) calibration Mössbauer absorption spectrum. The inset shows the scheme for the excitation of the  $\text{Fe}^{57}$  nucleus.

not taken into account by us here):

$$g_{00}^{\pi\pi} = g_{11}^{\pi\pi} = g_{10}^{\pi\pi} = g_{01}^{\pi\pi} = \frac{g_1}{v+i} + \frac{g_2}{v-v_0+i}, \quad (1)$$

$$g_{00}^{\sigma\sigma} = [1+3\cos^2(\varphi-\theta)] \frac{g_1}{v+i} + [1-\cos^2(\varphi-\theta)] \frac{g_2}{v-v_0+i},$$

$$g_{11}^{\sigma\sigma} = [1+3\cos^2(\varphi+\theta)] \frac{g_1}{v-i} + [1-\cos^2(\varphi+\theta)] \frac{g_2}{v-v_0+i}, \quad (2)$$

$$g_{01}^{\sigma\sigma} = g_{10}^{\sigma\sigma} = [\cos 2\theta + 3\cos(\varphi+\theta)\cos(\varphi-\theta)] \frac{g_1}{v+i} + [\cos 2\theta - \cos(\varphi+\theta)\cos(\varphi-\theta)] \frac{g_2}{v-v_0+i},$$

where  $g_1$  and  $g_2$  are the moduli of the resonant values of the coefficients respectively for the transitions  $\pm 1/2 \rightarrow \pm 1/2$  and  $\pm 1/2 \rightarrow \pm 3/2$  ( $g_1/g_2 = 1/3$ ,  $g_1 = 1.15 \cdot 10^{-5}$ ), while  $v_0$  is the distance between the resonances in units of  $\Gamma_n/2$  ( $v_0 = 7.8\Gamma_n/2$ ).

At a given orientation of the EFG axis, plane-polarized  $\pi$  and  $\sigma$  waves are the natural waves of the crystal. The input  $\pi$  wave excites a superposition of waves, and the same holds true also for  $\sigma$  waves. It is seen directly (1) that the necessary condition for the realization of suppression for the  $\pi$  superposition, namely vanishing of the parameter  $\Delta = g_{00}g_{11} - g_{01}g_{10}$ , is satisfied in the entire frequency range, including the two resonant frequencies. The physical nature of the equivalence of the resonance with respect to the SE for this polarization is connected with the fact at the employed orientation of the EFG axis, the  $\pi$  polarized wave can excite in the crystal only nuclear transitions with  $\Delta m = \pm 1$ . Therefore, in the first resonance there are excited simultaneously only two rather than four transitions, i.e., exactly as many as in the second resonance.

As to the  $\sigma$ -polarized wave, its magnetic wave vector has a component along the EFG axis, so that this wave is already capable of exciting in the first resonance, in addition to the transitions  $\Delta m = \pm 1$ , also the transitions  $\Delta m = 0$ , i.e., all four possible transitions. The polarization structure of the wave field in the transition to this resonance becomes strongly altered, and this leads immediately to violation of the SE conditions for the  $\sigma$  superposition:  $\Delta^\sigma(v=0) \neq 0$  [see (2)]. In the second resonance, the selection rules admit of only two tran-

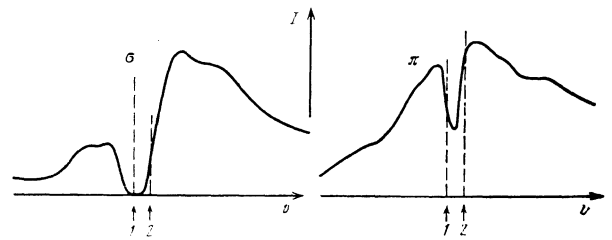


FIG. 6. Theoretical Mössbauer spectra of the (330) Laue diffraction, shown separately for polarized  $\pi$  and  $\sigma$  components of the  $\gamma$  beam.

sitions  $\Delta m = \pm 1$ , so that here the situation is analogous to the one considered above for the  $\pi$  superposition:  $\Delta^\sigma(v=v_0) = 0$ , and 100% SE is obtained. It is precisely the nonequivalence of the resonances with respect to the SE which causes the  $\sigma$ -polarized superposition to produce the experimentally observed drastic asymmetry in the passage of the gamma quanta through the crystal.

So far we have not dealt with the influence of the interference of transitions in different resonances on the overall picture. It is clear at the same time that by virtue of the smallness of the quadrupole splitting this influence can be quite strong. In the frequency interval between the resonances, the real parts of the first and second terms of the dynamic coefficient (1) and (2) have opposite signs, i.e., destructive interference takes place between the nuclear transitions. Strong compensation of the waves diffracted by the individual resonances takes place, and consequently the paired superposition state is destroyed. This explains the other distinguishing feature of the measured spectrum—the decrease of the intensity of the transmission of the quanta between the resonances. Obviously, the considered interference effect manifests itself principally in the passage, through the crystal, of the  $\pi$  superposition for which the SE takes place in both resonances.

Figure 6 shows separately the behavior of the  $\sigma$  and  $\pi$  polarizations, calculated by the KA theory. The emphasized features manifest themselves particularly clearly there: in the  $\sigma$  spectrum we have a strong asymmetry and complete lack of transparency of the crystal in a wide range of frequencies near the resonance on the  $\pm 1/2 \rightarrow \pm 1/2$  transition, while in the  $\pi$  spectrum we have high intensity of the transmission in both resonances and a radical decrease of the intensity in the intermediate region.

As to the large width of the obtained spectrum, it is the consequence, just as the previously observed broadening,<sup>1,6</sup> of the collective action of the crystal nuclei in the gamma-quantum scattering process (see Ref. 1). The wavelike profile of the experimental spectrum on the right wing (Fig. 4) is due to interference of the two paired states of the gamma quantum in the crystal.

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## Concentration dependence of local oscillations in Cu-Be system

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Inelastic scattering of cold neutrons was used to investigate the change of the phonon spectra of solid solutions of Be in Cu in the impurity concentration interval from 0.5 to 10 at.% with an aim at studying the transition from the local oscillations of a light isolated substitutional impurity to the impurity band. The measurements were made at room temperature. The experimental neutron-scattering cross sections were reduced in the noncoherent approximation. It was established that with increasing impurity concentration the following quantities vary linearly with the concentration: the energy position of the level of the local impurity oscillations, the width of this level, and the end-point frequency of the perturbed oscillation spectrum of the copper matrix.

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### INTRODUCTION

Local oscillations (LO) produced in the phonon spectrum following introduction of a light impurity atom, or of an atom more tightly bound to the atoms of the host crystal (matrix), have been under investigation for a long time. They were predicted theoretically back in 1942 in the studies of I. M. Lifshitz.<sup>1</sup> LO were first observed experimentally in ionic crystals by optical methods, and in metallic systems with the aid of the Mössbauer effect and of inelastic neutron scattering.<sup>2</sup> Naturally, the observed distributions of the spectral density of the LO are not the  $\delta$  functions predicted in the harmonic approximation for an isolated impurity. Nor is the LO lifetime, which is governed by the anharmonicity, the only factor that determines their real shape. Since any investigated system contains a finite impurity concentration, a definite role is played by the concentration broadening due to the interaction of the impurity atoms with one another.

However, there are practically no experimental investigations devoted to the concentration dependence of the LO width in metals where, unlike in ionic crystals, the restructuring of the electronic subsystem by introduction of an impurity influences substantially the character of the interaction between the atoms.

From among all the experimental methods of investigating impurity oscillations, inelastic scattering of slow neutrons is the most direct and most informative. The recently uncovered possibility of studying LO in

metallic systems by tunnel experiments presupposes prior knowledge of the electron-phonon interaction function  $\alpha^2(\omega)$ , and is consequently not a direct method.

The purpose of the present study was to trace, with the aid of inelastic scattering of cold neutrons, the onset of LO and their variation in a metallic system, using as an example Cu-Be alloys at concentrations that range from a practically isolated substitutional impurity to concentrations such that the interaction of the impurity centers of the perturbation cannot be neglected, and an LO band appears. Of definite interest here is the change of the phonon spectrum of the perturbed matrix.

### SAMPLES, MEASUREMENTS, AND DATA REDUCTION

The investigated Cu-Be system was chosen for a number of considerations. The dynamics of the copper lattice (matrix) has been sufficiently well investigated, as were also a number of physical properties of copper-beryllium alloys. Beryllium is an impurity with a single isotope. In addition, the LO due to the introduction of Be is as a rule far from the end point of the phonon spectrum of the matrix.<sup>3-5</sup>

The samples were made of specially purified copper containing not more than  $10^{-3}\%$  impurity. Binary copper alloys containing 0.5, 1, 2.7, 4.2, and 9.5 at.% Be, melted in vacuum, were homogenized at 1080 K for 10 hours and abruptly quenched to room temperature. The rolled sheets 1.5 mm thick were annealed in vacu-