# Resonant forward nuclear scattering of $\gamma$ rays after a stepped change in the energy of an excited nuclear state

Yu. V. Shvyd'ko, S. L. Popov, and G. V. Smirnov
*I. V. Kurchatov Institute of Atomic Energy, 123182, Moscow*

(Submitted 24 January 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **53**, No. 4, 217–223 (25 February 1991)

The forward rescattering of $\gamma$ rays by an ensemble of resonant nuclei after reversal of the hyperfine magnetic field has been studied. The experimental data show that the energy of the intermediate excitation in the system of nuclei is changed by the field reversal. The excitation decays spontaneously, with a coherent enhancement of the radiative channel and a forward emission of $\gamma$ rays at a shifted frequency.

The coherent scattering of a $\gamma$ ray in a system of identical nuclei involves the formation of an intermediate excited state, which propagates to the entire nuclear system. In this state, there is a certain probability amplitude for each of the nuclei to be excited. In other words, there is a space–time correlation among the amplitudes and phases of the nuclear excitations in the ensemble of nuclei.[1] Until recently, the coherence effects characteristic of the decay of collective excited states of this sort have been studied only in the course of elastic scattering of $\gamma$ rays.[2–4] It has recently been shown that the correlation in the amplitudes and phases of the nuclear excitations in the excited state and thus the coherence effects also prevail in an inelastic resonant scattering of $\gamma$ rays, specifically, during steady-state ultrasonic excitation of a nuclear ensemble.[5]

In the present letter we are reporting a study carried out to determine whether the excitation in a nuclear ensemble remains a collective excitation if the energy of the intermediate excited state of the nuclei is changed quickly by a time-varying external perturbation ("quickly" here means that the change occurs in a time $\tau \ll \tau_0$, where $\tau_0 = 1/\Gamma_0$ is the natural lifetime of the nucleus). Indications of the retention of the space-time correlation in the phases of the nuclear excitations should be *a directed emission* of $\gamma$ rays and *an enhancement of the radiative decay channel.*[1] If the excitation is localized, the reradiation of $\gamma$ rays should be isotropic, and the decay should result in the usual $\gamma$ yield, e.g., about 11% in the decay of the first excited state of the $^{57}$Fe nucleus. As we know, a localization occurs during a scattering accompanied by a change in the ground-state energy of the nucleus[6] (as a result of a flip of the nuclear spin in the hyperfine magnetic field).

In this study we examined the scattering of $\gamma$ rays in the original direction, which is one of the directions along which a coherent reradiation of $\gamma$ rays can occur during the decay of a collective nuclear excitation. The experimental apparatus is shown schematically in Fig. 1. We studied the scattering of $^{57}$Fe nuclei in a matrix of the

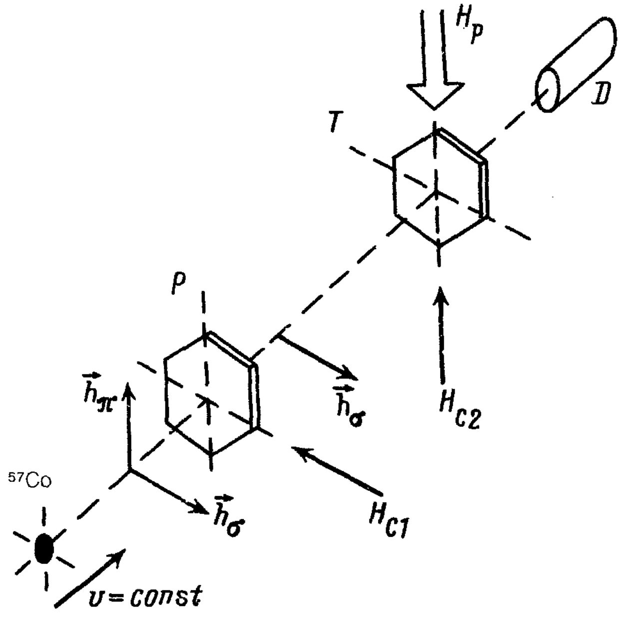

FIG. 1. Experimental layout. $^{57}$Co—$^{57}$Co(Cr) source of 14.4-keV $\gamma$ rays; *P*—polarizer ($^{57}$FeBO$_3$); *T*—target ($^{57}$FeBO$_3$); *D*—NaI(Tl) $\gamma$ detector.

magnetic crystal $^{57}FeBO_3$ (the target, $T$), enriched to 95% in the resonant isotope. Another $^{57}FeBO_3$ crystal served as a polarizer ($P$). The $^{57}Co(Cr)$ Mössbauer source (with a linewidth $\Gamma_s = 0.2$ mm/s) was moved at a constant velocity $v$ in such a way that the $\gamma$-ray emission at the frequency $\omega_s$ selectively excited one of the transitions between the ground state and the first excited state of the nuclei in the $^{57}FeBO_3$ crystals, where the corresponding energy levels were split by the hyperfine interaction. The polarizer crystal had a thickness $L_p = 30$ $\mu$m, which corresponds to a very large resonant-absorption factor, $\mu L_p \simeq 84$ (for a transition width $\Delta m = 0$). The crystal was initially put in a saturated (single-domain) state by an applied magnetic field $\vec{H}_{c1} = 30$ Oe. It transmitted essentially one polarization component of the radiation (the $\vec{h}_\sigma$ component in the case of a transition with $\Delta m = 0$). The target crystal was also in a saturated state because of an applied magnetic field $\vec{H}_{c2} = 6$ Oe ($\vec{H}_{c2} \perp \vec{H}_{c1}$). The nuclei interacted with the component of the radiation selected by the polarizer. The target had a thickness $L_T = 17$ $\mu$m ($\mu L_T \simeq 48$ for $\Delta m = 0$). The system of two magnetized $^{57}FeBO_3$ crystals was thus originally opaque.

The energy of the nuclear states was varied by putting the nuclei in another hyperfine sublevel, by reversing the hyperfine magnetic field (Fig. 2, a and b). The field was reversed through a (180°) magnetization reversal of the target $^{57}FeBO_3$ crystal ($T$). This magnetization reversal resulted from the application of a magnetic field $\vec{H}_p = 20$ Oe $\gg \vec{H}_{c2}$, antiparallel to $\vec{H}_{c2}$, at the time $t = 0$. The reversal occurred over a time $\tau_s \leqslant 5$ ns, which is much shorter than $\tau_0$ and also much shorter than the period of the Larmor spin precession of the $^{57}Fe$ nucleus. The magnetization-reversal time was short because of the magnetic properties and quality of the $^{57}FeBO_3$ crystal.[7,8] The reversed state was maintained for 400 ns. The reversal of the magnetic field was repeated every 2 $\mu$s. The time evolution of the intensity of the $\gamma$-ray emission from the target after the 180° magnetization reversal was measured. The $\gamma$ emission was detected by a method similar to that described in Refs. 8 and 9. The detector moni-

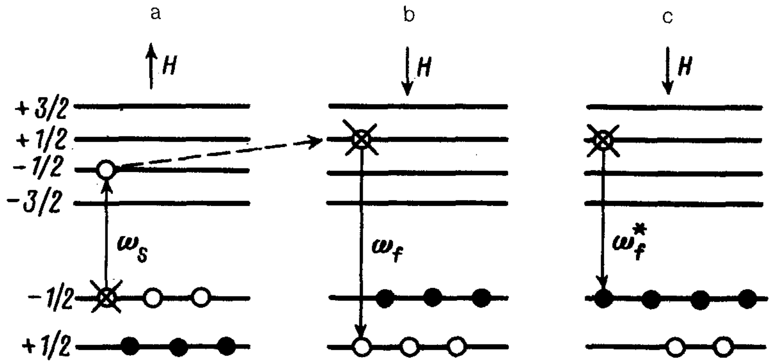

FIG. 2. Energy-level diagram of $^{57}Fe$ and level populations (a) before and (b,c) after the reversal of the hyperfine magnetic field. Parts b and c show two possible mechanisms for the decay of the excited nuclear state.

tored an angle $\sim 10^{-5}$ sr, so the emission scattered in the original direction was detected.

We turn now to the experimental results. Figure 3 shows the time evolution of the intensity of the $\gamma$-ray emission from the target in the original direction. The time $t = 0$ corresponds to the beginning of a magnetization reversal. Each time dependence was measured during the selective excitation, by $\gamma$ radiation with a frequency $\omega_s$, of one of the hyperfine-structure transition in the $^{57}FeBO_3$ crystal (Fig. 3, a–c), with a natural frequency $\omega_i$ ($i = 1,2,3$; see the right side of Fig. 3). In all three cases, an intense short burst of $\gamma$-ray emission was observed behind the target after its magnetization reversal. This burst reached and even slightly exceeded the intensity of the radiation incident on the target (the dashed line in this figure). We recall that before the magnetization reversal the target was an essentially black absorber for resonant $\gamma$ rays. In two cases

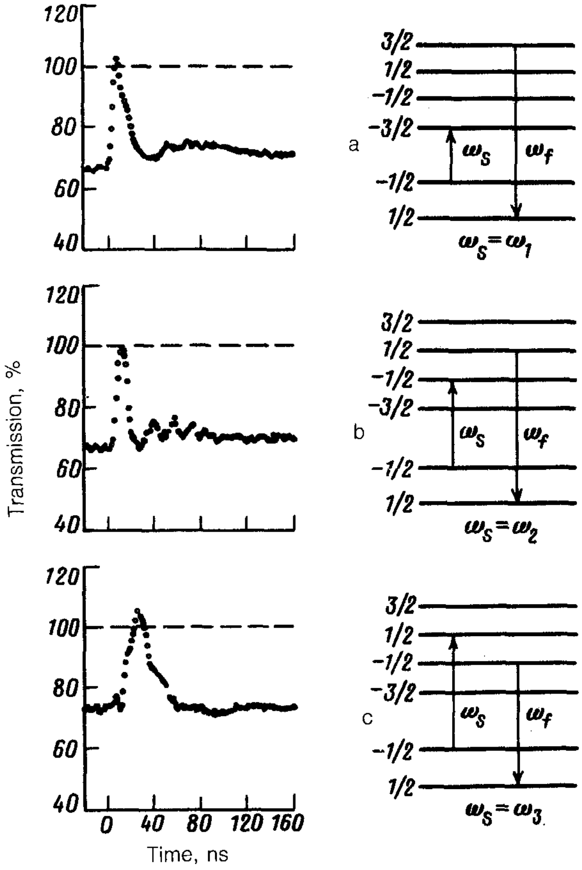

FIG. 3. Time evolution of the intensity of the $\gamma$ radiation from the $^{57}FeBO_3$ Mössbauer target in the original direction, after a reversal (at $t = 0$) of the hyperfine magnetic field a the nuclei. The nuclear hyperfine-structure transitions in the $^{57}FeBO_3$ crystal with different natural frequencies $\omega_i$ ($i = 1,2,3$) were excited selectively.

(Fig. 3, a and b), a weak and more prolonged second burst appeared after the end of the first burst. Intensity beats were clearly observed against the background of this second burst.

The curves of the time evolution which were found have approximately the same overall shape, but they do differ in detail: (I) The peak of the first burst is reached at different times during the excitation of different transitions, earliest in case *a* and latest in case *c*. (II) The bursts differ in length, being shortest in case *b* and longest in case *c*. (III) The beat frequency in case *a* is higher than that in case *b*.

Figure 4 shows the results of more-detailed studies of the time evolution during the excitation of the nuclear transition $-1/2 \Rightarrow -1/2$ (case *b* in Fig. 3). The curves measured at different energies of the incident radiation near the nuclear resonance $\omega_s = \omega_2 \pm 3\Gamma_0$ differ from each other in the intensity of the first $\gamma$ burst. The height of the burst increases sharply as the resonance is crossed in the direction of increasing energy. In the case $\omega_s = \omega_2 + 3\Gamma_0$ the height of the burst is noticeably above the intensity of the incident radiation, and its intensity is twice that of the $\gamma$ radiation absorbed by the target before the magnetic field reversal. The intensity beats are of approximately the same frequency on all three curves.

Let us analyze the results. Let us take a more detailed look at the processes which might determine the measured time evolution. Figure 2 shows the energy-level diagram of the nuclei before and after the reversal of the hyperfine magnetic field. The filled and open circles represent subsystems of nuclei ("black" and "white" nuclei, respectively) which are in different sublevels of the ground state. Each sublevel is characterized by a definite projection of the nuclear spin. If there is an approximately equal filling of the sublevels at room temperature, only half the nuclei participate in the interaction with the incident radiation. As an example we selected the situation in which the radiation incident on the target,[1] $\mathscr{E}\exp(-i\omega_s t)$ excites the

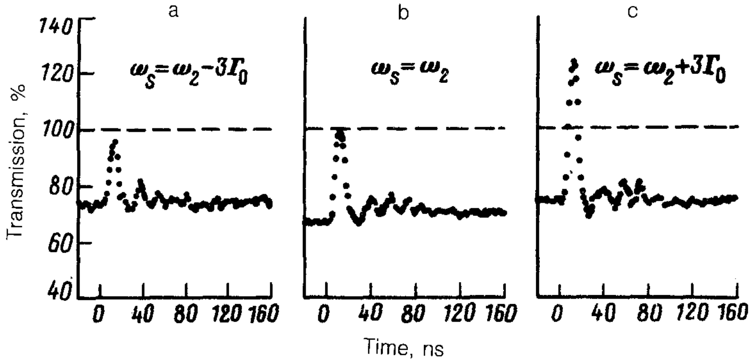

FIG. 4. Time evolution of the intensity of the $\gamma$ radiation from the $^{57}$FeBO$_3$ Mössbauer target in the original direction after the reversal (at $t=0$) of the hyperfine magnetic field at the nuclei. The $-1/2 \rightarrow -1/2$ transition, with the natural frequency $\omega_2$, was excited. b—Exactly at resonance; a,c—in the wings of the resonance.

$-1/2 \rightarrow -1/2$ transition, i.e., interacts with the subsystem of "white" nuclei. After the flip of the nuclear spins in the hyperfine magnetic field, the energies of the nuclei in both the excited state and the ground state change. Under the conditions which result, the incident radiation begins to interact with the subsystem of black nuclei. Since the time scale for the excitation of the nuclei is on the order of $\tau_0$, the target remains partially transparent to the incident radiation over this time. In other words, a wave $\mathscr{E}\mathscr{F}_1(t)\exp(-i\omega_s t)$, where $\mathscr{F}_1(t)$ is a function that decays in time $\mathscr{F}_1(0) = 1$, begins to pass through the target at $t = 0$.

The subsystem of white nuclei, on the other hand, escapes from the influence of the incident radiation and begins to decay spontaneously. This decay can go by a conversion channel or a radiative channel. In the radiative channel, the selection rules allow a transition to the ground state accompanied by the emission of a $\gamma$ ray with a frequency $\omega_f$ or $\omega_f^*$ (Fig. 2, b and c). The final state in the first of these cases is identical to the initial state; the original number of nuclei remains in each subsystem. In the latter case, in contrast, one white nucleus becomes black, so a marker appears in the system of nuclei signaling a localization of the excitation in the course of the scattering. The coherence is evidently disrupted, as in the case of a decay by the conversion channel. It would appear at first glance that the probabilities for the incoherent decay channels might be quite high, but actually they are not. We will return to this matter below. Consequently, one can speak of a preservation of the collective nature of the intermediate excitation of nuclei only in the case of a resonant scattering as in parts *a* and *b* of Fig. 2. Which of these situations is realized experimentally?

We begin by playing out the case in which there is a complete loss of the space–time correlation among the phases of the nuclear excitations upon a stepped change in the energy. This event may occur either as a result of a localization of the excitation or as a result of an unsynchronized reversal of the hyperfine field at the various nuclei. The secondary $\gamma$ radiation would be isotropic, and it would amount to a negligible fraction of the radiation at the detector. Only the radiation at the frequency $\omega_s$, $\mathscr{E}\mathscr{F}_1(t)\exp(-i\omega_s t)$, would reach the detector. If the phase correlation is disrupted, regardless of which transition is excited, the intensity beyond the target would reach its maximum at the same time—at the time of the field reversal. This maximum intensity would be $\mathscr{E}^2$. In addition, the time evolution would not have any intensity beats. We see that this model does not correspond to the experimental results, so we can discard it.

We now assume that the magnetization reversal does not disrupt the phase correlation of the nuclear excitations that arise in the system of white nuclei upon the capture of the exciting $\gamma$ ray. In this case the spontaneous decay of the collective excitation should be of a coherent nature. By this we mean that there should be an elevated probability for the emission of a $\gamma$ ray, at the frequency $\omega_f$, during the decay. In addition, the emission of the $\gamma$ ray should occur predominantly forward.[10] The probability for the emission of an internal-conversion electron, like the probability for the emission of a photon with a frequency $\omega_f^*$, should be small with respect to the probability for the coherent process.

Consequently, along with the transmitted wave $\mathscr{E}\mathscr{F}_1(t)\exp(-i\omega_s t)$ a wave with the frequency $\omega_f$, $\mathscr{E}\mathscr{F}_2(t)\exp(-i\omega_f t)$, appears beyond the target as a result of the reradiation by the nuclei. An interference of the wave packets with the different carrier frequencies, $\omega_s$ and $\omega_f$, should be observed. One can show that in the case of a very thick nuclear target ($\mu L_{\rm T} \gg 1$), excited precisely at resonance ($\omega_s = \omega_i$), we should have $\mathscr{F}_2(t) = -\mathscr{F}_1(t) = -J_0(\sqrt{\mu L_{\rm T}/\tau_0})$. The resultant intensity of the $\gamma$ radiation in the original direction should then have a time dependence.

$$I(t) = 4\mathscr{E}^2 J_0^2(\sqrt{\mu L_{\rm T} t/\tau_0}) \sin^2[(\omega_s - \omega_f)t/2]. \qquad (1)$$

In other words, this model tells us that intensity beats modulated by a Bessel function should be observed experimentally. Let us analyze expression (1).

At the time $t = 0$ the interfering waves are out of phase, and they cancel each other out: The intensity is zero. At a certain time $t_i$ the intensity $I(t)$ reaches a maximum and then takes the form of beats in time with a period $T_i = 2\pi|\omega_s - \omega_f|$. It follows that the period of the beats and the time at which the intensity reaches its maximum are different and are determined by the frequency difference $\omega_s - \omega_f$ in the case in which nuclear transitions with different natural frequencies $\omega_i (i = 1,2,3)$ are excited. The values $t_2 - t_1 = 2$ ns and $t_3 - t_1 = 16$ ns found from (1) agree with the experimental results ($3.5 \pm 1.2$ ns and $17.5 \pm 1.2$ ns, respectively). The calculated values of the beat period, $T_1 = 8.8$ ns and $T_2 = 15.1$ ns, again correspond to the experimental results ($T_1 = 8.2$ ns and $T_2 = 15.5$ ns). For the $i = 3$ transition the calculated value is $T_3 = 55.5$ ns, comparable to the length of the transients, so beats with such a period probably could not be observed. The duration of the first burst is determined by the Bessel function which reflects the absorption and reradiation of the $\gamma$ ray in a thick target. As the resonant absorption factor $\mu L_{\rm T}$ becomes larger, the first burst becomes shorter. For example, we find $\mu_2 L_{\rm T} = 48$ (Fig. 3b) and $\mu_3 L_{\rm T} = 12$ (Fig. 3c).

As we move away from the resonance, the reradiated wave will have an initial phase which depends on the difference $\omega_s - \omega_i$. This circumstance will alter the conditions for the interference of the transmitted and rescattered waves. This circumstance determines the dependence of the intensity of the first burst on the frequency of the exciting $\gamma$ radiation (Fig. 4).

It can be concluded from the set of experimental data that a fast reversal of the hyperfine magnetic field and the resulting transition of the nuclei to a new energy state in the course of the nuclear resonant scattering of $\gamma$ rays do not destroy the phase correlation among the nuclear excitations in the intermediate state. The transition of the nuclei to a different energy sublevel would make it possible to see a spontaneous decay of the intermediate excited state. The high intensity of the burst of $\gamma$-ray emission after the reversal (Fig. 4c) is evidence that the amplitudes of the incident and scattered waves are comparable. This result in turn is evidence of a *directionality of the reradiation* or $\gamma$ rays and a *coherent enhancement of the radiative channel* during the spontaneous decay of the intermediate excited state.

[1)] This model-based analysis is restricted to only a single Fourier component of the radiation.

---

[1] A. M. Afanas'ev and Yu. Kagan, Pis'ma Zh. Eksp. Teor. Fiz. 2, 164 (1965) [JETP Lett. 2, 104 (1965)].

[2] G. V. Smirnov, Hyperfine Interaction **27**, 203 (1986); U. Van Buerck, Hyperfine Interactions **27**, 219 (1986).

[3] Yu. V. Shvyd'ko and G. V. Smirnov, J. Phys. Cond. Matter **1**, 10563 (1989).

[4] R. Rueffer, E. Gerdau, M. Grote, R. Hollatz, R. Roehlsberger, H. D. Rueter, and W. Sturhahn, Hyperfine Interactions **61**, 1279 (1990).

[5] S. L. Popov, G. V. Smirnov, and Yu. V. Shvyd'ko, Pis'ma Zh. Eksp. Teor. Fiz. **49**, 651 (1989) [JETP Lett. **49**, 747 (1989)]; S. L. Popov, G. V. Smirnov, and Yu. V. Shvyd'ko, Hyperfine Interactions **58**, 2463 (1990).

[6] A. N. Artem'ev, V. V. Sklyarevskiĭ, G. V. Smirnov, and E. P. Stepanov, Zh. Eksp. Teor. Fiz. **63**, 1390 (1972) [Sov. Phys. JETP **36**, 736 (1973)].

[7] O. S. Kolotov, V. A. Pogozhev, G. V. Smirnov, Yu. V. Shvyd'ko, S. Kadeckova, M. Kotrbova, and J. Novak, J. Phys. Status Solidi a **72**, k197–k201 (1982).

[8] G. V. Smirnov, Yu. V. Shvyd'ko, O. S. Kolotov, V. A. Pogozhev, M. Kotrbova, S. Kadeckova, and J. Novak, Zh. Eksp. Teor. Fiz. **86**, 1495 (1984) [Sov. Phys. JETP **59**, 875 (1984)].

[9] G. V. Smirnov, Yu. V. Shvyd'ko, É. Realo, Pis'ma Zh. Eksp. Teor. Fiz. **39**, 33 (1984) [JETP Lett. **39**, 41 (1984)]; G. V. Shvyd'ko, Pis'ma Zh. Eksp. Teor. Fiz. **44**, 431 (1986) [JETP Lett. **44**, 556 (1986)].

[10] Yu. V. Shvyd'ko, G. V. Smirnov, S. L. Popov, and T. Hertrich, Pis'ma Zh. Eksp. Teor. Fiz. **53**, 69 (1991) [JETP Lett. **53**, 69 (1991)].

Translated by D. Parsons