Controlling Absorption of Gamma Radiation via Nuclear Level Anticrossing

R. Coussement,[1,2,3] Y. Rostovtsev,[1] J. Odeurs,[2] G. Neyens,[2] H. Muramatsu,[2] S. Gheysen,[2] R. Callens,[2] K. Vyvey,[2] G. Kozyreff,[3] P. Mandel,[3] R. Shakhmuratov,[2,4] and O. Kocharovskaya[1,5]

[1]*Department of Physics and Institute for Quantum Studies, Texas A&M University, College Station, Texas 77843-4242*
[2]*Instituut voor Kern en Stralingsfysica, Katholieke Universiteit Leuven, Celestijnenlaan 200 D, B-3001 Leuven, Belgium*
[3]*Optique Nonlinéaire Théorique, Université Libre de Bruxelles, Campus Plaine CP 231, Bld du Triomphe, B-1050 Bruxelles, Belgium*
[4]*Kazan Physico-Technical Institute of Russian Academy of Sciences, 10/7 Sibirsky trakt, Kazan 420029, Russia*
[5]*Institute of Applied Physics, Russian Academy of Sciences, 46 Ulyanov Street, 603300 Nizhny Novgorod, Russia*

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A significant reduction of absorption for single gamma photons has been experimentally observed by studying Mössbauer spectra of ^{57}Fe in a $FeCO_3$ crystal. The experimental results have been explained in terms of a quantum interference effect involving nuclear level anticrossing due to the presence of a combined magnetic dipole and electric quadrupole interaction.

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Electromagnetically induced transparency (EIT) [1,2] has been successfully demonstrated under different experimental conditions: in continuous wave and pulsed regimes [3], with atomic and molecular gases (at room temperature [4,5] or at low temperature [6]), with solids doped by rare-earth ions [7] and semiconductor quantum wells [8], for different wavelengths ranging from optics to microwaves [3,4,9]. Although it is more difficult to deal with gamma radiation than with optical radiation [10], a few coherent effects for gamma rays have been predicted. Some of them have already been demonstrated, such as the photon echo via stepwise phase modulation of recoilless gamma radiation [11], Rabi flopping by microwave driving of hyperfine transitions [12,13], storage of nuclear excitation via magnetic switching [14], reversed time [15] and dynamical beating [16] in Mössbauer spectra, gamma-microwave double resonance [13,17,18], or gamma-optical double resonance [19]. Recently, interesting proposals have been discussed to obtain lasing for gamma rays by utilizing coherent effects [20–23]. In this Letter, we report on experiments demonstrating the EIT effect at the single-photon level via the level (anti)crossing technique. A theory of the one-photon interaction with a nucleus has been developed to describe the experimental results. The obtained results open an interesting perspective to extend coherent effects to nuclear transitions.

Figure 1 represents the main results of the paper. It shows the observed Mössbauer spectrum of a single crystal of $FeCO_3$ at a temperature of 30.5(5) K which corresponds to a magnetic hyperfine field of $B = 15.1(3)$ T. At this field the hyperfine levels $|m = 1/2\rangle$ and $|m = -3/2\rangle$ anticross. For the transitions connected to the anticrossing levels, a deficit of absorption of 25% is observed at the peak velocity. It means that some transparency is induced by interference, similar to EIT observed in optics.

The experiments were performed by using a conventional Mössbauer setup. It includes a source of gamma radiation (^{57}Co<u>Rh</u>), an absorber of $FeCO_3$ cleaved on the $\{1014\}$ faces (optical thickness is of the order 10), and a detector. The absorber was mounted on a target holder which allows for a precise temperature control at the target position in the interval 4–600 K. Besides the magnetic hyperfine field, the Fe^{2+} nucleus in the $FeCO_3$ crystal [24,25] is subjected to a large axially symmetric electric field gradient (EFG) which results in a well-resolved quadrupole doublet. The level structures of the source and the absorber are shown in Fig. 2. In a magnetic field, the levels might shift to the position where their energies coincide; this situation is referred to as *level crossing.* But due to the presence of additional fields, the energies of levels might never be equal, and it is the case of *level anticrossing.*

If the magnetic field is collinear with the EFG axis, the axial symmetry is preserved and the m states are eigenfunctions of the total nuclear Hamiltonian if the z axis is chosen along the symmetry axis. However, in such a mineral containing impurities and defects, one can expect a small distribution of fields which are responsible for the

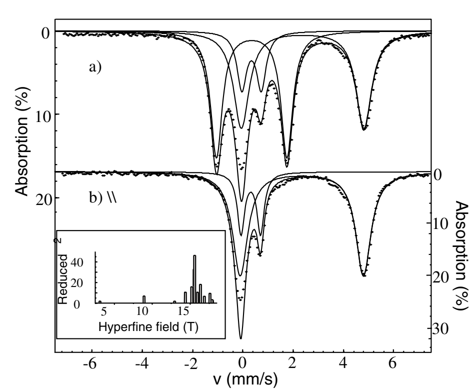

FIG. 1. Observed Mössbauer spectra (dots) from a single crystal of $FeCO_3$ in the (a) perpendicular and (b) parallel geometry. The solid curves are the spectra obtained when the coherence and interference effects are ignored. The inset gives the χ^2 of all fits as a function of the hyperfine field.

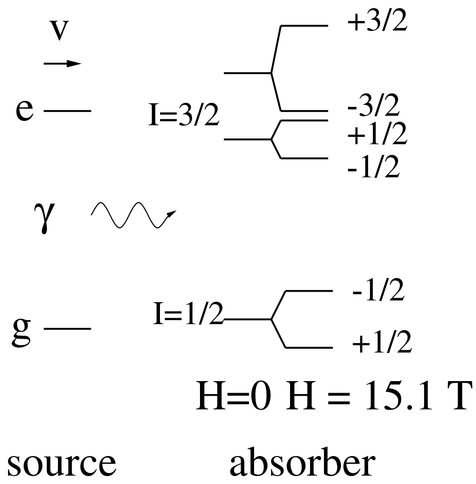

FIG. 2. The level structure of source and absorber.

observed inhomogeneous broadening and for a weak breaking of the axial symmetry. This symmetry breaking is so small that it can be neglected except for two levels that are very close in energy as it happens for two crossing levels. For those levels the small breaking of the axial symmetry results in a coupling between different m levels. This coupling mixes the m levels and allows for interference in the gamma transitions. The static Hamiltonian of the absorber can be written as a sum of three terms,

$$\hat{H}_A = \hat{H}_Q + \hat{H}_{B_{||}} + \hat{H}_{B\perp}, \tag{1}$$

where $\hat{H}_Q$ describes the quadrupole interaction, $\hat{H}_Q = \hbar\Omega_Q(3\hat{I}_z^2 - \hat{I}^2)$, $\Omega_Q = eQV_{zz}/4\hbar I(2I-1)$; Q is the quadrupole moment of the nucleus; $\hat{H}_{B_{||}} = \Omega_{B_{||}}\hat{I}_z$ describes the Zeeman interaction due to the magnetic field component along the EFG axis; $\hat{H}_{B\perp}$ is the term that breaks the axial symmetry—it could be either a Zeeman component ($\hat{H}_{B\perp} = -\Omega_{B\perp}\hat{I}_x$) or an asymmetry of the EFG; and $\Omega_B = g\mu B/\hbar$ is the Larmor frequency at which the nucleus rotates in the magnetic field. Note that in a frame rotating with this Larmor precession the effect of the magnetic field component $B_{||}$ is compensated by the Coriolis term. Consequently, in that rotating frame, we are left with a quadrupole interaction, and the coupling term now acts as an ac driving field [26] that resonantly couples the crossing levels $m = -3/2$ and $m = 1/2$. Such a scheme is similar to those in optics which show EIT.

The absorber $FeCO_3$ is ferromagnetic below 38.3 K, which means that the hyperfine field can be varied by changing the temperature. Analyzing a series of Mössbauer spectra taken at different temperatures, we determine that at 30.5(5) K the states $|I = 3/2, m = -3/2\rangle$ and $|I = 3/2, m = +1/2\rangle$ cross and are mixed by the coupling term $\hat{H}_{B\perp}$ (Fig. 2). Two of the absorption lines ($m = -1/2 \leftrightarrow m = -3/2$) and ($m = -1/2 \leftrightarrow m = 1/2$) merge into one single line. For this line a deficit in absorption has been observed experimentally. Let us note that the six transitions, which are allowed between the excited and ground states, can be represented in the form of three doublets. Every doublet consists of two lines. Each line can be obtained from the doublet partner by reversing the direction of the z axis ($m \to -m$). Because of equal populations in the ground state and because of the fundamental chiral symmetry of electromagnetic interactions, doublet partners must have the same shape, the same width, and the same amplitude. In the analysis of the experimental data this doublet structure gives a constraint on the number of free parameters. All spectra can be fitted very well putting that constraint except the spectra at the crossing field where a deficit in absorption is observed (see the inset of Fig. 1). Note that the observed effects are substantially different from those due to the interference between resonant and photoelectric absorption, which occurs for narrow Mössbauer lines at lower energies [27].

The deficit in absorption can be explained as an interference of the two transition amplitudes corresponding to the two lines which occur at the same energy. Such a coherence effect is known in quantum optics as EIT. This phenomenon is possible only if the involved m states are not pure, but coupled. In our experimental configuration the coupling must be caused by an interaction breaking the axial symmetry, such as a perpendicular component of the magnetic hyperfine field with respect to the EFG axis, or by an asymmetry in the quadrupole interaction. This interaction most probably originates from crystal imperfections and impurities. The latter are well known to cause small distributions of the electromagnetic fields. They are, e.g., observed in the inhomogeneous broadening of the Mössbauer lines. They can, in addition, induce a weak breaking of the axial symmetry leading to an anticrossing of the levels due to a coupling between the $|I = 3/2, m = -3/2\rangle$ and $|I = 3/2, m = +1/2\rangle$ states and, hence, cause EIT.

Obviously, in order to conclude that we have observed EIT, we need to rule out the other possible causes for reduced absorption. When two absorption lines overlap and a thick target has been used, the total intensity of the coinciding lines is not necessarily the sum of the intensities of the two partner lines due to saturation effects. Therefore, one can still argue that effective thickness effects are the origin of the observed deficit. The problem can be addressed in two ways. First, it can be proved by simulations using the Maxwell-Bloch equations that the change in effective thickness cannot explain the reduced absorption. Here we show the results of another experiment which was designed such that thickness effects are totally excluded. In this experiment we observed the Mössbauer spectra with the gamma radiation parallel rather than perpendicular to the hyperfine axis. The parallel geometry, contrary to the perpendicular geometry, rules out possible thickness effects as explained below.

The source does not experience hyperfine fields. The emitted photons will have a well defined circular polarization, either $\sigma = +1$ or $\sigma = -1$, each occurring statistically with the same probability. In general, each photon can induce different Δm transitions according to the transformation of the photon wave function when rotating from the propagation axis to the axis of the hyperfine fields:

$$|\sigma\rangle = \sum_{\Delta m} D^1_{\Delta m,\sigma}(\theta)|\Delta m\rangle. \tag{2}$$

In the perpendicular geometry ($\theta = \pi/2$) each photon has three possible polarizations, $\Delta m = 1$, $\Delta m = 0$, and $\Delta m = -1$, whatever its circular polarization. It makes that the effective thickness at the sum peak can be changed giving some correction for the total intensity. We know, however, from simulations that this correction is too small to explain the observed effect. For the parallel geometry the situation is even clearer. In this case $\theta = 0$ and, hence, $\Delta m = \sigma$, and only one transition can be induced by a photon, either $\Delta m = 1$ or $\Delta m = -1$, depending statistically on the circular polarization. Therefore, the effective thicknesses of such a transition and its doublet partner are exactly equal. Consequently, the intensity of the sum peak is expected to be the sum of the intensities of the two partners. This is not what is observed experimentally. Any departure in the experimental spectrum from this sum rule can be due to only coherence and interference effects. The solid line in Fig. 1 is obtained as a sum of two lines which have the same shape, width, and intensity as their respective partner lines. There is clearly a strong experimental deficit in absorption, or transparency, which we will now identify as similar to EIT in optics.

In order to consider the coherent effects induced by single gamma photons, the quantum description of radiation must be employed. First, the quantum field created by the source is calculated. Then, this field is used to calculate the absorption coefficient for the absorber. The Hamiltonian of the source is

$$\hat{H}_S = \epsilon_a|e\rangle\langle e| + \epsilon_b|g\rangle\langle g| + \hat{H}_{\text{bath}}, \tag{3}$$

where $\hat{H}_{\text{bath}} = \hbar\sum_k(g_k b_k|e\rangle\langle g| + g_k b_k^+|g\rangle\langle e|)$ accounts for the relaxation, $g_k = \mathcal{P}_{eg}/\hbar\sqrt{\hbar\omega_k/2\epsilon_0 V}$ is the coupling coefficient, $\mathcal{P}_{eg}$ is the dipole moment of the transition, V is the quantization volume, ω_k is the frequency of the field mode having wave number k, and b_k and b_k^+ are photon annihilation and creation operators. The corresponding field operator [28] is

$$\hat{E} = \sum_k \frac{g_k \exp(i\omega_k t - i\vec{k}\vec{r})}{\omega_k - \omega_{eg} + i\gamma_S}\hat{b}_k^+, \tag{4}$$

where $\omega_{eg} = \omega_e - \omega_g$ and $\gamma_S = \omega_{eg}^3\mathcal{P}_{eg}^2/3\pi\epsilon_0 c^3\hbar^2$ is the relaxation of the nuclear coherence.

Absorption occurs if the photon frequency is resonant with the corresponding transition of the absorber. Depending on the direction of the magnetic field with respect to the propagation direction of the gamma photons, different numbers of resonances are observed. Six lines are observed for an emission perpendicular to the z axis and four lines for an emission parallel to the z axis. For the particular magnetic field at which the levels cross, two lines coincide for each configuration. At this field, anticrossing instead of crossing occurs if the axial symmetry is broken. These anticrossing levels are the main focus of our analysis. In order to gain physical insight in the system we will consider a model system that involves only these anticrossing levels, as shown in Fig. 3(a). Levels $|a\rangle$ and $|c\rangle$ are coupled by the interaction that breaks the axial symmetry and, hence, anticross. Level $|b\rangle$ is the ground state of the system. Transitions $a \leftrightarrow b$ and $c \leftrightarrow b$ have different polarizations. Hence, we introduce two reservoirs to describe relaxation with different polarizations. The Hamiltonian of the simplified model of the absorber is

$$\hat{H}_A = \hat{H}_0 + \hat{H}_f + \hat{H}^{ab}_{\text{bath}} + \hat{H}^{cb}_{\text{bath}}, \tag{5}$$

where $H_0 = \epsilon_a|a\rangle\langle a| + \epsilon_b|b\rangle\langle b| + \epsilon_c|c\rangle\langle c|$, and

$$\hat{H}_f = \hbar\hat{\mathcal{E}}|a\rangle\langle b| + \hbar\hat{\mathcal{E}}^+|b\rangle\langle a| + \hbar\Omega|c\rangle\langle a| + \hbar\Omega^*|a\rangle\langle c|. \tag{6}$$

$\hat{H}^{ab}_{\text{bath}}$ and $\hat{H}^{cb}_{\text{bath}}$ are the interactions with the reservoirs to describe relaxation via spontaneous emission. The energies ϵ_a and ϵ_c depend on the longitudinal component of the magnetic field, $\hat{\mathcal{E}} = \mathcal{P}_{eg}\hat{E}/\hbar$, and Ω is the coupling between levels $|a\rangle$ and $|c\rangle$. Assuming that initially all the population is in $|b\rangle$ and the photon field Rabi frequency is small compared to the relaxation rate, the upper states of the absorber will remain nearly unpopulated. The simplified equations for the atomic and field operators can be written in the form (see [28], Chaps. 7 and 12, and [29])

$$\dot{\hat{\sigma}}_{ab} = -\Gamma_{ab}\hat{\sigma}_{ab} - i\hat{E} - i\Omega\hat{\sigma}_{cb} + \hat{F}_{ab}, \tag{7}$$

$$\dot{\hat{\sigma}}_{cb} = -\Gamma_{cb}\hat{\sigma}_{cb} - i\Omega^*\hat{\sigma}_{ab} + \hat{F}_{cb}, \tag{8}$$

where $\Gamma_{ab} = \gamma + i\omega_{ab}$, $\Gamma_{cb} = \gamma + i\omega_{cb}$, γ is the relaxation rate of the nuclear coherence of the absorber, $\hat{\sigma}_{ij} = \sum_n |i\rangle\langle j|^{(n)}/N$, $\hat{F}_{ab}$ and $\hat{F}_{cb}$ are the usual Langevin forces

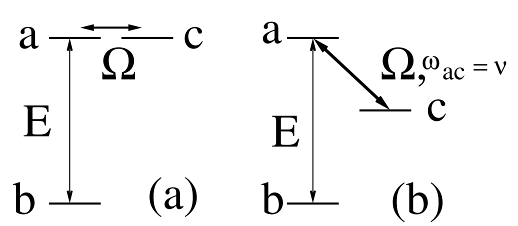

FIG. 3. (a) Static frame. Simplified level structure of level anticrossing: the misalignment component of the magnetic field (providing coupling Ω between levels a and c) acts as a driving field in optical schemes [28]. (b) Rotating frame. The coupling static field in the rotating frame acts like an ac field with the frequency equal to transition frequency. The level scheme is resembling the Λ scheme in optics.

with $\langle \hat{F}_{ij} \rangle = 0$, and $\langle \hat{E}\hat{F}_{ij} \rangle = \langle \hat{F}_{ij}\hat{E} \rangle = \langle \hat{E}^{+}\hat{F}_{ij} \rangle = \langle \hat{F}_{ij}\hat{E}^{+} \rangle = 0$. Note that the axial symmetry breaking interaction acts as the driving field in a three-level Λ system [1]. Equations (7) and (8) for the present scheme are similar to the ones in the Λ scheme [2]. The essential difference between both schemes is the driving field which in its present scheme is static [30], while in the Λ one it is periodic. Furthermore, one can transform the present scheme in a Λ scheme (and vice versa) by changing from a static reference frame to a rotating one. From these similarities we can term the observed transparency as EIT for a single gamma photon. The absorption coefficient of gamma radiation, $D = \langle \dot{n}_a \rangle = i\langle \hat{E}^{+}\sigma_{ab} - \sigma_{ba}\hat{E} \rangle$, is proportional to the rate of excitation of state $|a\rangle$ by the probe field $\hat{E}$ and equals $\Re[\Gamma/(\Gamma^2 + |\Omega|^2)]$, where $\Gamma = \gamma + \gamma_S + ikv$ and $\omega_{eg} = \omega_{ab}$. Finally, the absorption profile for single photons has a similar form as in the case of coherent field driving. The Maxwell-Bloch calculations for the system produce results similar to the experimental spectra.

This creates interesting perspectives to extend the coherent effects already known for atomic media to nuclei. For example, one interesting feature of the EIT resonance is the steep dispersion producing a time delay via an ultraslow group velocity [31]. We expect that the group velocity of gamma rays can be considerably reduced at the anticrossing. According to an estimation based on the Maxwell-Bloch equations ($v_g = 8\pi|\Omega|^2/3\lambda^2 N\gamma \simeq 10^5$ cm/s), the delay of the gamma rays in a thick sample could be of the order of 100 ns which is 4 orders of magnitude longer than without anticrossing. The delay depends upon the magnitude of the symmetry breaking interaction (for example, the interaction can be caused by a misaligned component of the magnetic field), and it could be experimentally observed.

In conclusion, the EIT effect has been observed for single gamma photons. The effect can be explained by the anticrossing of two m states due to the presence of a combined magnetic dipole and electric quadrupole interaction which are not fully collinear.

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