Nonreciprocity of Gamma Emission and Absorption due to Quantum Coherence at Nuclear-Level Crossings

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We show that the presence of destructive interference due to quantum coherence at nuclear Zeeman level crossings can totally preclude γ-resonant absorption under appropriate conditions leaving resonant emission intact. The underlying principle can be demonstrated in a Mössbauer effect experiment and has broad consequences in the domain of gamma optics.

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Coherence in radiation is generally observed when a single photon simultaneously excites two levels, and the two levels emit a photon leaving the system (of atomic or nuclear spins) in the same final state. In principle, the arrangement consists of two quantum paths linked through a pair of intermediate levels $|1\rangle$ and $|2\rangle$, which start from the initial level $|i\rangle$ and end on the same final level $|f\rangle$ as shown in Fig. 1(a). The energy spacing of the pair of levels $|1\rangle$ and $|2\rangle$ must be less than the sum of the radiative and homogeneous widths for these to be excited simultaneously by a single photon. Angular momentum conservation would require the photon transition from $|i\rangle$ to $|1\rangle$ and $|2\rangle$ to be allowed by the atomic or nuclear transition and the photon polarization selection rules with similar considerations valid for the second photon absorption from $|1\rangle$ and $|2\rangle$ to $|f\rangle$. In such a case, novel quantum coherent effects can manifest if the amplitudes a_{i1} (a_{1f}) and a_{i2} (a_{2f}) for the first (second) photon transitions can interfere destructively or constructively. In atomic systems, these basic ideas have led to the phenomenon of emission without absorption and have stimulated interest to realize lasers without inversion [1–3], darkline resonances [4], and refractive index enhancement [5]. The corresponding nuclear case has remained largely unexplored, however. It is the purpose of this Letter to show that nonreciprocity between resonant emission and absorption of γ rays in a crystal can occur, and such an observation could serve as a basis to realize a γ-ray laser. At the outset it may be stated that nonreciprocity between emission and absorption in either an atomic or a nuclear spin system is the result of a different density matrix in a ground state from that in an excited state, as we shall show in this work. It is not related to any violation of a fundamental symmetry law.

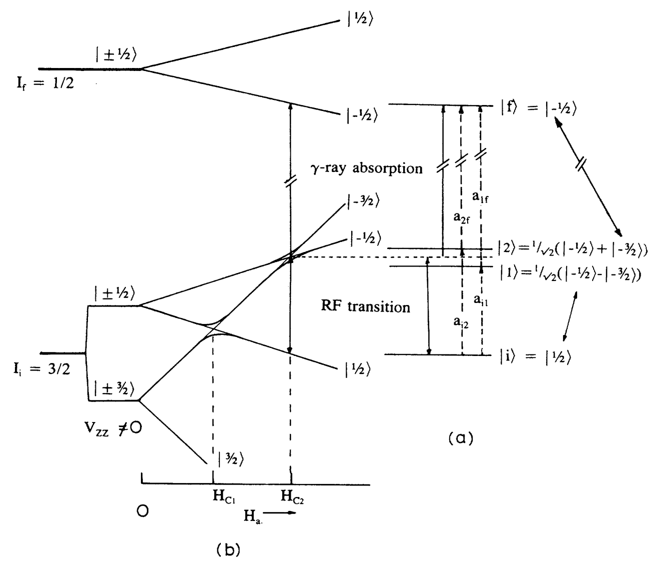

FIG. 1. (a) γ-ray quantum interference in absorption observed between the doublet $|1\rangle$ and $|2\rangle$ and the final level $|f\rangle$, when an rf field is applied between the initial level $|i\rangle$ and the doublet. The two quantum paths between $|i\rangle$ and $|f\rangle$ through the doublet are shown by broken lines while the corresponding photon transitions are shown in bold lines. In practice, the doublet is produced by nuclear level crossings of Zeeman levels, as illustrated in (b) for a nucleus of spin $I_i = \frac{3}{2}$ in the ground state. Presence of an electric field gradient and a magnetic field leads to level crossings at H_{c1} and H_{c2}.

To illustrate the principle of the present method consider a nucleus with ground- and excited-state spins of $I_i = \frac{3}{2}$, $I_f = \frac{1}{2}$, respectively. Consider these nuclei to reside in a noncubic uniaxial crystal, which is immersed in an external magnetic field $\mathbf{H}_a$ applied parallel to the c axis of the crystal. The underlying hyperfine level splittings are shown in Fig. 1(b). In the noncubic crystalline host the presence of a nuclear quadrupole interaction will lead to a splitting of the $I_i = \frac{3}{2}$ ground level (since $I_i > \frac{1}{2}, eQ \neq 0$) into a doublet. The magnetic field produces a Zeeman splitting which is linear in H_a. This results in two nuclear-level crossings at fields H_{c_1} and H_{c_2} as shown in Fig. 1(b).

It is well known [6,7] that if the applied magnetic field $\mathbf{H}_a$ is tilted by a small angle β with respect to the electric field gradient (V_{zz}) axis, then the pair of Zeeman levels

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$|-\frac{3}{2}\rangle$ and $|-\frac{1}{2}\rangle$ will strongly mix, producing a doublet with an energy spacing (at H_{c_2}) of $2\langle-\frac{3}{2}|H_\perp|-\frac{1}{2}\rangle = \omega_B \sin\beta\langle-\frac{3}{2}|I_x|-\frac{1}{2}\rangle$, where $H_\perp$ represents the axial symmetry breaking part of the Zeeman Hamiltonian and $\hbar\omega_B = g\mu_n H_\parallel$. The level scheme of Fig. 1(b) at H_{c_2} bears a close analogy to that of Fig. 1(a); level $|i\rangle$ represents the $|m=+\frac{1}{2}\rangle$ level of the ground state, the pair of intermediate levels $|1\rangle$ and $|2\rangle$ represents the two mixed nuclear levels which are a superposition of the pure Zeeman states $|m=-\frac{1}{2}\rangle$ and $|m=-\frac{3}{2}\rangle$, while the final state $|f\rangle$ represents the Zeeman level $|m=-\frac{1}{2}\rangle$ of the nuclear existed state $|I_f=\frac{1}{2}\rangle$.

Coherence between the pair of levels $|1\rangle$ and $|2\rangle$ can be achieved by using a single rf photon to simultaneously excite the doublet starting from the level $|i\rangle$, provided the rf bandwidth exceeds the energy splitting of the pair, and the transitions $|i\rangle \rightarrow |1\rangle$ and $|i\rangle \rightarrow |2\rangle$ are allowed for rf absorption. This process will produce a density matrix $(\rho_{11},\rho_{22},\rho_{12})$ that contains coherent terms like ρ_{12} that we shall calculate later. Absorption of a γ ray from such a coherently prepared ensemble of nuclear spins will in general show quantum interference between the amplitudes a_{1f} and a_{2f}. We can calculate the γ-ray absorption rate $(d\rho_{ff}/dt)$ in terms of the density matrix terms $(\rho_{11}, \rho_{22},\rho_{12})$ using the Von Neumann equations generally used to describe the time evolution of a system

$$\frac{\partial\rho_{11}}{\partial t} = (i/\hbar)(\rho_{1f}H_{f1} - H_{1f}\rho_{f1}) - \Gamma\rho_{11}\,, \tag{1}$$

$$\frac{\partial\rho_{22}}{\partial t} = (i/\hbar)(\rho_{2f}H_{f2} - H_{2f}\rho_{f2}) - \Gamma\rho_{22}\,, \tag{2}$$

$$\frac{\partial\rho_{ff}}{\partial t} = -(i/\hbar)(\rho_{1f}H_{f1} - H_{1f}\rho_{f1}) - (i/\hbar)(\rho_{2f}H_{f2} - H_{2f}\rho_{f2})\,, \tag{3}$$

$$\frac{\partial\rho_{1f}}{\partial t} = -(i\omega_{1f} + \tfrac{1}{2}\Gamma)\rho_{1f} + (i/\hbar)\rho_{12}H_{2f} + (i/\hbar)(\rho_{11} - \rho_{ff})H_{1f}\,, \tag{4}$$

$$\frac{\partial\rho_{2f}}{\partial t} = -(i\omega_{2f} + \tfrac{1}{2}\Gamma)\rho_{2f} + (i/\hbar)\rho_{21}H_{1f} + (i/\hbar)(\rho_{22} - \rho_{ff})H_{2f}\,, \tag{5}$$

$$\frac{\partial\rho_{12}}{\partial t} = -(i\omega_{12} + \Gamma)\rho_{12} + (i/\hbar)(\rho_{1f}H_{f2} - H_{1f}\rho_{f2})\,, \tag{6}$$

where $H_{ab} = W_{ab}e^{-i\omega t}$, with W_{ab} representing the nuclear and photon matrix element for the transition $|a\rangle$ to $|b\rangle$, Γ is the radiative width for nuclear resonant absorption, and $\delta_{1(2)}$ is the detuning of the γ-ray energy from levels $|1(2)\rangle$.

Solution of these equations can be obtained by integrating Eqs. (4) and (5) and replacing ρ_{11}, ρ_{22}, and ρ_{12} by their initial values ρ_{11}^0, ρ_{22}^0, and ρ_{12}^0. The general solution is

$$\frac{\partial\rho_{ff}}{\partial t} = (W_{1f}{}^2/4\hbar^2)[\Gamma/(\Gamma^2/4+\delta_1^2)](\rho_{11}^0 - \rho_{ff}^0) + (W_{2f}{}^2/4\hbar^2)[\Gamma/(\Gamma^2/4+\delta_2^2)](\rho_{22}^0 - \rho_{ff}^0) + [(W_{2f}W_{1f})/4\hbar^2]\left[\left(\frac{\Gamma + i\omega_{12}}{(\Gamma/2 - i\delta_1)(\Gamma/2 + i\delta_2)}\right)\rho_{12}^0 + \text{c.c.}\right]. \tag{7}$$

For emission of a γ ray resulting from the transitions $|f\rangle \rightarrow |1\rangle$ and $|f\rangle \rightarrow |2\rangle$, the initial conditions are $\rho_{ff}^0 = 1$ and ρ_{11}^0, ρ_{22}^0, $\rho_{12}^0 = 0$. From Eq. (7), the transition rate reduces to the sum of two Lorentzians

$$-\frac{\partial\rho_{ff}}{\partial t} = (W_{1f}{}^2/4\hbar^2)\Gamma/(\Gamma^2/4+\delta_1^2) + (W_{2f}{}^2/4\hbar^2)\Gamma/(\Gamma^2/4+\delta_2^2)\,. \tag{8}$$

For absorption of a γ ray, on the other hand, the initial conditions require ρ_{11}^0, ρ_{22}^0, ρ_{12}^0 to be each finite and $\rho_{ff}^0 = 0$, and from Eq. (7), the transition rate now includes an interference term in addition to the two Lorentzians, i.e.,

$$\frac{\partial\rho_{ff}}{\partial t} = (W_{1f}{}^2/4\hbar^2)[\Gamma/(\Gamma^2/4+\delta_1^2)]\rho_{11}^0 + (W_{2f}{}^2/4\hbar^2)[\Gamma/(\Gamma^2/4+\delta_2^2)]\rho_{22}^0 + [(W_{2f}W_{1f})/4\hbar^2]\left[\frac{\Gamma + i\omega_{12}}{(\Gamma/2 - i\delta_1)(\Gamma/2 + i\delta_2)}\rho_{12}^0 + \text{c.c.}\right]. \tag{9}$$

It is of interest to identify at least some of the conditions under which the interference term of Eq. (9) completely cancels the contribution of the two Lorentzians to give a vanishing absorption rate, i.e., $\partial\rho_{ff}/\partial t \rightarrow 0$. To accomplish this, first of all we need to calculate the initial density matrix of the pair of levels $|1\rangle$ and $|2\rangle$ when an rf field is turned on to couple these levels to the initial level $|i\rangle = |m=\frac{1}{2}\rangle$. This is achieved using the Liouville equations

$$\frac{\partial\rho_{11}}{\partial t} = -\Gamma\rho_{11} - \frac{2\Gamma H_{\text{rf}}^2|V_{1i}|^2}{(\Gamma^2+\Delta_1^2)\hbar^2}(\rho_{11} - \rho_{ii})\,, \tag{10}$$

$$\frac{\partial\rho_{22}}{\partial t} = -\Gamma\rho_{11} - \frac{2\Gamma H_{\text{rf}}^2|V_{2i}|^2}{(\Gamma^2+\Delta_2^2)\hbar^2}(\rho_{22} - \rho_{ii})\,, \tag{11}$$

$$\frac{\partial\rho_{12}}{\partial t}=-(i\omega_{12}+\Gamma)\rho_{12}-\frac{H_{\rm rf}^2V_{1i}V_{2i}}{\hbar^2}\left[\frac{\rho_{11}-\rho_{ii}}{\Gamma-i\Delta_1}+\frac{\rho_{22}-\rho_{ii}}{\Gamma+i\Delta_2}\right],\tag{12}$$

$$\frac{\partial\rho_{ii}}{\partial t}=-\Gamma\rho_{ii}+\frac{2\Gamma H_{\rm rf}^2|V_{1i}|^2}{(\Gamma^2+\Delta_1^2)\hbar^2}(\rho_{11}-\rho_{ii})+\frac{2\Gamma H_{\rm rf}^2|V_{2i}|^2}{(\Gamma^2+\Delta_2^2)\hbar^2}(\rho_{22}-\rho_{ii}),\tag{13}$$

where $H_{\rm rf}$ and Ω represent the coupling strength and frequency of an rf field, $V_{ji}=\langle j|I_++I_-|i\rangle$ represents the angular matrix element of an rf transition between Zeeman states $|j\rangle$ and $|i\rangle$, while $\Delta_1=\Omega-\omega_1$ and $\Delta_2=\Omega-\omega_2$ represent detuning of the rf field. To obtain the steady state solution when a continuous rf field is applied, we equate $\partial\rho_{11}/\partial t=\partial\rho_{22}/\partial t=\partial\rho_{12}/\partial t=0$ and obtain the density matrices

$$\rho_{11}^0=\frac{2|C_1|^2}{1+2|C_1|^2}\rho_{ii}^0,\quad \rho_{22}^0=\frac{2|C_2|^2}{1+2|C_2|^2}\rho_{ii}^0,\tag{14}$$

$$\rho_{12}^0=\frac{V_{1i}V_{i2}}{\Gamma+i\omega_{12}}\frac{H_{\rm rf}^2}{\hbar^2}\left[\frac{1}{1+2|C_1|^2}\frac{1}{\Gamma-i\Delta_1}+\frac{1}{1+2|C_2|^2}\frac{1}{\Gamma+i\Delta_2}\right]\rho_{ii}^0,\tag{15}$$

where

$$C_1=\frac{V_{1i}}{\hbar}\frac{H_{\rm rf}}{\Gamma-i\Delta_1}\quad\text{and}\quad C_2^*=\frac{V_{i2}}{\hbar}\frac{H_{\rm rf}}{\Gamma+i\Delta_2}.$$

For the case when the rf frequency (Ω) is tuned halfway in between levels $|1\rangle$ and $|2\rangle$ it is easy to show that detunings $\Delta_1=-\Delta_2=-\frac{1}{2}\omega_{12}$. Further if we assume $V_{1i}=\pm V_{i2}$, then $C_1=\pm C_2^*$ and the γ-absorption rate becomes

$$\frac{\partial\rho_{ff}}{\partial t}\propto\frac{|W_{1f}|^2}{4\hbar^2}\frac{\Gamma}{\Gamma^2/4+\delta_1^2}+\frac{|W_{2f}|^2}{4\hbar^2}\frac{\Gamma}{\Gamma^2/4+\delta_2^2}\pm\frac{W_{2f}W_{f1}}{4\hbar^2}\left[\frac{\Gamma-i(\omega_{12}/2)}{(\Gamma/2-i\delta_1)(\Gamma/2+i\delta_2)}+\text{c.c.}\right].\tag{16}$$

In Eq. (16), δ_1 and δ_2 represent the γ-ray detuning from the anticrossing point, i.e., $\delta_{1(2)}=\omega_{1(2)f}-\omega$.

If one considers a γ-ray absorption event starting midway between levels $|1\rangle$ and $|2\rangle$ and ending on the final state $|f\rangle=|m=-\frac{1}{2}\rangle$, it follows that the detunings $\delta_1=-\delta_2=\frac{1}{2}\omega_{12}$. Furthermore, if the conditions

$$W_{1f}=\mp W_{2f}\quad\text{and}\quad V_{1i}=\pm V_{2i}\tag{17}$$

are satisfied, it then follows from Eq. (16) that the γ-absorption rate will vanish identically bringing us to the central point of this Letter. Under these circumstances the interference term [third term in Eq. (16)] is equal to and opposite in sign to the sum of the first two Lorentzian terms leading to a complete cancellation of terms. Thus complete quantum destructive interference, i.e., switching off the nuclear resonant absorption, occurs and this dramatic effect can be demonstrated in a Mössbauer effect experiment. In emission such an effect is excluded because as shown earlier in Eq. (8), the interference term is absent.

It is appropriate to inquire under what conditions can a complete quantum destructive interference effect be realized in the laboratory for known nuclear-level spin sequence encountered in Mössbauer spectroscopy. For the spin sequence $\frac{3}{2}\rightarrow\frac{1}{2}$ shown in Fig. 1(b), if an rf field is tuned between $|i\rangle$ and the pair of levels $|1\rangle$ and $|2\rangle$, the selection rule for rf transitions ($\Delta m=\pm1$) will only mix the $|i\rangle=|m=\frac{1}{2}\rangle$ state with the $|m=-\frac{1}{2}\rangle$ component of the $|1\rangle$ and $|2\rangle$ pair of levels. In this case the rf matrix elements V_{1i}, V_{2i} are equal in magnitude and sign. Now if a γ-resonant absorption experiment is performed in the longitudinal geometry, i.e., $\gamma\|\mathbf{H}_a$, then it is well known that the $\Delta m=0$ components are suppressed while the $\Delta m=\pm1$ components are allowed. Then, γ-ray absorption from the $|1\rangle$ and $|2\rangle$ pair of levels to the nuclear excited level $|f\rangle=|m=-\frac{1}{2}\rangle$, can only couple to the $|m=-\frac{3}{2}\rangle$ component of the pair, and therefore $W_{1i}=-W_{2i}$. We thus recognize that the conditions of Eq. (17) are completely fulfilled and therefore one can expect the γ-resonant absorption to be completely suppressed for the condition $\delta_1=-\delta_2$. The situation discussed above is by no means unique and there are other experimental schemes that can be realized where complete quantum destructive interference effect can be observed.

In summary, we have for the first time described the principle and method to observe γ-ray quantum interference effects. Specifically, we have identified the conditions under which a complete suppression of nuclear resonant absorption can occur while leaving resonant emission intact. An application of this concept resides in γ-ray lasers [8] where suppression of absorption will significantly ease the pumping and gain requirements for lasing to occur. The underlying nonreciprocity, we believe, also has profound consequences to demonstrate γ-ray optical effects, such as polarization filters and large refractive index change.

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