Coherent Optical Control of Mössbauer Spectra

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A compound nuclear-electron system driven by a coherent optical field and probed at the gamma-ray Mössbauer transition is analyzed for the first time. The possibility of optical control of the Mössbauer spectra is predicted. This study provides the basis for a new branch of spectroscopy, laser-Mössbauer spectroscopy, and opens prospects for the realization of a gamma-ray laser. [S0031-9007(99)09013-4]

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Coherently driven, three-level atoms are widely used in quantum optics. Recently, novel effects have been observed that are caused by the excitation of atomic coherence. Examples of such effects include lasing without inversion, electromagnetically induced transparency, and high refractive index with vanishing losses [1].

Similar effects could be seen in the gamma-ray range if it were possible to drive nuclear transitions coherently. However, as is well known, direct coherent driving of nuclear transitions is not feasible. Polarization of nuclei using microwave radiation is extremely ineffective because the populations of hyperfine sublevels are nearly equal, even at liquid helium temperature. Optical driving of a nuclear transition is not efficacious because the energies of the photons are typically orders of magnitude away from those corresponding to the nuclear transitions. X-ray sources for resonant coherent driving of nuclei are unavailable, and gamma-ray lasers do not exist at all. Although some indirect ways of using laser fields (via multiphoton processes or near-field effects) to manipulate gamma-ray transitions have been discussed, they are extremely low efficiency methods and hard to implement experimentally [2].

In this Letter, we show that coherent optical driving of an electron transition can drastically modify the absorption-emission spectra for gamma-ray transitions due to the hyperfine coupling between the nuclear and electronic degrees of freedom. The hyperfine interaction has been extensively studied both in nuclear and electronic spectra, and widely used in different kinds of spectroscopy technique (such as nuclear magnetic resonance, double radio-frequency optical resonance, coherent population trapping, etc.) as well as for laser separation of isomers [3]. It was considered also [4,5] the possibility to perturb the Mössbauer transition by a resonant microwave driving of Zeeman sublevels in the strong magnetic field at low temperature ($\mu_B B > kT$). However, the idea of a coherent optical control of Mössbauer spectra, as far as we know, has not been discussed.

Model of the compound system: nucleus and electron.—The study of coherent optical control of the gamma-ray Mössbauer spectra requires a consideration of *the compound nuclear-electron system* coupled with both optical drive and gamma-ray probe fields. The simplest model of the compound system must involve at least four energy levels: $|1\rangle = |G, g\rangle$, $|2\rangle = |G, e\rangle$, $|3\rangle = |E, g\rangle$, $|4\rangle = |E, e\rangle$. Here $|G\rangle$ and $|E\rangle$ stand for the ground and excited states of a nucleus and accordingly $|g\rangle$ and $|e\rangle$ stand for the ground and excited states of an electron. In the absence of the hyperfine interaction, two pairs of transitions, 1-2 and 3-4, as well as 1-3 and 2-4, are exactly degenerate (Fig. 1a). The hyperfine interaction leaves them nearly degenerate, because of its relative weakness. For the same reason, the selection rules remain essentially the same as in the absence of the interaction. Hence the transitions may change the electronic state or the nuclear state, but not both. This compound system is driven by the coherent optical field at the 1-2 and 3-4 transitions, and probed by gamma rays at the 1-3 and 2-4 transitions. We note that such a doubly degenerate four-level system is rather unusual both for nuclear and electronic transitions separately, but appears naturally in the compound system.

The Hamiltonian of the system has the form $\hat{H} = \hat{H}_0 + \hat{V}$, where $\hat{H}_0$ is the Hamiltonian of the free atom $\hat{H}_0 = \hbar\omega_1|1\rangle\langle 1| + \hbar\omega_2|2\rangle\langle 2| + \hbar\omega_3|3\rangle\langle 3| + \hbar\omega_4|4\rangle\langle 4|$. The coupling $\hat{V}$ describes the interaction of the atom with the drive and probe fields

$$\hat{V} = \hbar\Omega(|1\rangle\langle 2| + |3\rangle\langle 4|) + \hbar\alpha(|1\rangle\langle 3| + |2\rangle\langle 4|) + \text{H.c.},$$

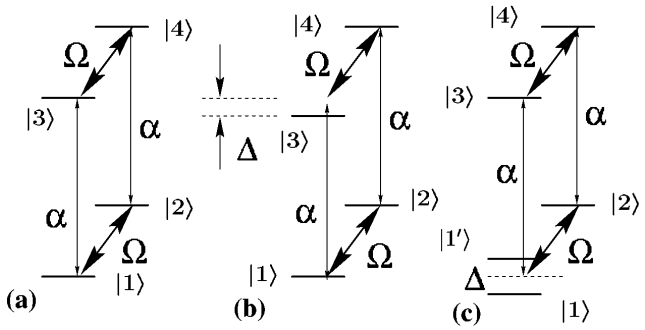

FIG. 1. Compound nuclear-electron system in the absence of hyperfine interaction (a), and in the presence of the hyperfine interactions, leading to the shift (b) and the splitting (c) of an energy level.

where $\Omega = \mu_e \mathcal{E}_0/2\hbar$, $\alpha = \mu_n \mathcal{E}/2\hbar$ are the Rabi frequencies of the drive and probe transitions; μ_e, μ_n, $\mathcal{E}_0$, $\mathcal{E}$ are the effective moments and fields at the electronic and γ-ray transitions.

Master equations.—The common master equations involving only diagonal relaxation terms cannot be directly applied here. The correct master equations are

$$\begin{aligned}
\dot{\sigma}_{13} &= -\Gamma_{13}\sigma_{13} + \gamma_0\sigma_{24} + i\alpha n_{13} + i\Omega^*\sigma_{14} - i\Omega\sigma_{23}\,,\\
\dot{\sigma}_{24} &= -\Gamma_{24}\sigma_{24} + i\alpha n_{24} - i\Omega^*\sigma_{14} + i\Omega\sigma_{23}\,,\\
\dot{\sigma}_{12} &= -\Gamma_{12}\sigma_{12} + \gamma\sigma_{34} + i\Omega n_{12} + i\alpha^*\sigma_{14} - i\alpha\sigma_{32}\,,\\
\dot{\sigma}_{34} &= -\Gamma_{34}\sigma_{34} + i\Omega n_{34} - i\alpha^*\sigma_{14} + i\Omega\sigma_{32}\,.
\end{aligned} \tag{1}$$

They involve the cross-relaxation terms ($\gamma_0\sigma_{24}$, $\gamma\sigma_{34}$) appearing as a result of degeneracy of the transitions. Here $n_{ij} = n_i - n_j$ is a population difference between states $|i\rangle$ and $|j\rangle$, γ_0^{-1} and γ^{-1} are the lifetimes of populations on the optical and nuclear transition, respectively, $\Gamma_{ij} = \gamma_{ij} + i(\omega - \omega_{ji})$, γ_{ij} and ω_{ji} are the line broadening and frequency for the transition $i \leftrightarrow j$. The equations for the coherences σ_{23} and σ_{14}, and populations have a standard form. Similar cross-relaxation terms have been introduced earlier in [6] to handle for the degeneracy of magnetic sublevels in the excited and ground electronic state. They play a very important role for the compound nuclear electron system. In particular, because of these terms, the coherent optical driving of the electrons does not affect the gamma-ray nuclear transitions in the completely symmetrical degenerate case (see Fig. 1a).

The hyperfine interaction between the nucleus and electron arises from several different mechanisms: Coulomb interaction of the nuclear and electron charges, the magnetic dipole interaction of the nuclear spin and total momentum of the electron, and the quadrupole interaction. The hyperfine interaction breaks the symmetry of the doubly degenerate four-level scheme by shift, Δ, and splitting of one or more energy levels (Figs. 1b and 1c).

The typical value of this asymmetry is of the order of 10^{-7}–10^{-6} eV. Hence, the asymmetry is always small on the scale of both optical and gamma-ray transitions. However, in the case when this value exceeds the widths of both transitions, this asymmetry allows for the *resonant* selective optical driving of only one of the two (or of many in case of multiple splittings) optical transitions either in the ground or excited state of the nucleus. We therefore predict very efficient modification of both populations and coherences in the compound nuclear-electron system. Moreover, a sufficiently strong optical drive can essentially modify the Mössbauer spectra even in the case of broad optical lines ($\gamma_0 \gg \Delta$). These important conclusions do not depend on the specific mechanism of the hyperfine interaction.

Frequency shift.—Let us consider a simple model for the frequency shift of state 3 (Fig. 1b): the isomer shift of the electron ground S state for the excited-state nucleus, due to a change of its radius, as compared to the ground-state nucleus.

The absorption coefficient is defined by $k = \mathrm{Im}(P/\mathcal{E})$, where $P = \mu_n \mathcal{N}(\sigma_{13} + \sigma_{24})$ is the polarization, $\mathcal{N}$ is the density of atoms. The absorption spectra, calculated on the basis of Eqs. (1), are depicted in Fig. 2. The Mössbauer spectrum without optical driving is presented in Fig. 2a, and consists of one line (transition 1-3). A driving field resonant with the transition 1-2 populates level 2. Hence, the additional line, corresponding to the transition 2-4, which is shifted from 1-3 by the magnitude of the frequency shift, appears in the spectrum as soon as the drive is applied (Fig. 2b).

Increasing the driving-field intensity (Fig. 2c) leads to the Rabi splitting of transitions 1-3 and 2-4 (the Autler-Townes effect). Moreover, an electromagnetically induced transparency (vanishing absorption) on the transition 1-3 appears. Let us note that the lines of the nuclear transitions are broadened due to the coupling with the optical transition (whose width is assumed to be larger than the width of the gamma-ray transition).

A further increase of the driving-field intensity, i.e., Rabi frequency ($\Omega \gg \Delta$) leads to a narrowing of the absorption lines toward the natural width (Fig. 2d). This

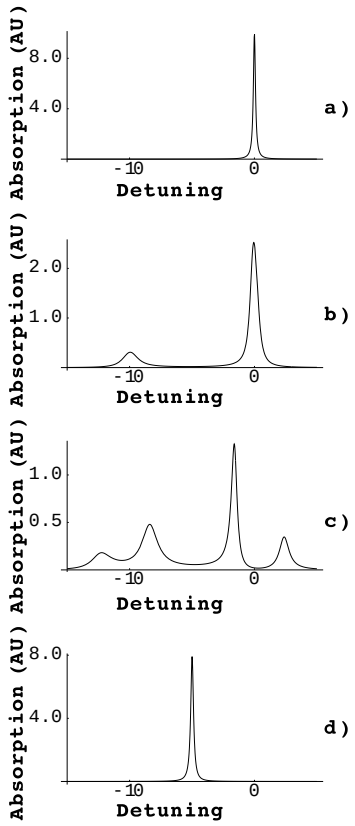

FIG. 2. Modification of the Mössbauer absorption spectra for the scheme involving frequency shift (Fig. 1b) with increase of the intensity of the driving field: (a) $\Omega = 0$; (b) $\Omega = 0.3\gamma_0$; (c) $\Omega = 2.0\gamma_0$; (d) $\Omega = 100\gamma_0$. The common parameters are $\Delta = 10\gamma_0$, $\gamma = 0.2\gamma_0$.

is caused by the effective decoupling of the electron and nuclear subsystems, due to the strong driving of both 1-2 and 3-4 transitions (when the Rabi frequency exceeds the frequency shift). This phenomenon arises from the atomic interference caused by the cross relaxation at the transitions 1-3 and 2-4. Let us also note that, in the limit of a strong field, the lines corresponding to the Rabi satellites shift far away and become weak, while a central line appears shifted with respect to its original position. This last line shift is a result of the residual hyperfine interaction. Its origin is clear in the dressed states picture. In the limit of a strong field, the dressed-state frequencies for the ground nuclear state are $\pm\Omega$, while for the excited nuclear state due to the presence of the detuning one has shifted dressed-state frequencies $-\Delta/2 \pm \Omega$. Thus, the central line is shifted by $\Delta/2$. The same result follows from the equation for polarization in the limit $\Omega \gg \Delta$, $\mathcal{P} = i\alpha\mu_n\mathcal{N}(n_{13} + n_{24})/[\gamma/2 + i(\omega - \omega_{31} - \frac{\Delta}{2})]$. Clearly, the line shift is exactly equal to $\Delta/2$, and the line width is the same as in the absence of driving, $\gamma/2$. Note that the frequency shift is preserved even when the width of the optical transition exceeds the magnitude of this shift.

Other schemes.—We have also analyzed four different other model schemes involving the splitting of any of the four energy levels into two sublevels. Effects similar to those described above appear in these schemes. Some additional effects were found in the system with the lowest level $|1\rangle$ split into two sublevels (Fig. 1c). The most obvious one is an optical pumping of atoms into a sublevel that is not coupled to a driving field. This results in a polarization of the nuclear spin and hence in vanishing absorption from the sublevel that is depleted by optical pumping. In the case of a monochromatic field driving the 1-2 and $1'$-2 transitions simultaneously, both 1-3 and $1'$-3 absorption lines practically disappear as a result of coherent population trapping (Fig. 3). This means that the medium becomes transparent for the gamma-ray radiation. This result is preserved even when both the optical and γ-ray lines overlap the hyperfine structure.

Similar effects can be also observed in the emission-absorption spectra of the excited nuclei. Resonant optical driving of the electronic transition in the excited nuclear state provides also some additional features such as the appearance of new emission lines as a result of Raman scattering. Adding an incoherent pump to the resonant optical drive produces the typical schemes of inversionless amplification in the gamma-ray range such as V, Λ, and double Λ (which have been realized in optics [7]).

Laser-Mössbauer spectroscopy.—The following modifications of Mössbauer absorption-emission spectra under the action of coherent optical driving are possible. (1) Appearance of new Mössbauer lines (shifted with respect to original lines by the magnitude of the hyperfine structure). (2) Dynamic Stark splitting of both original and additional Mössbauer lines (Autler-Townes effect). (3) Broadening

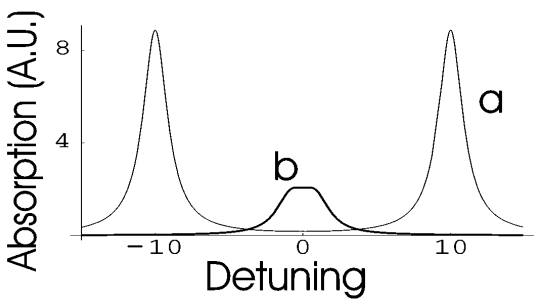

FIG. 3. Modification of the Mössbauer absorption spectra for the scheme involving level splitting with increase of the intensity of the driving field: (a) $\Omega = 0$; (b) $\Omega = 100\gamma_0$.

of the Mössbauer lines caused by the fast relaxation at the optical transition. (4) Vanishing of some Mössbauer lines and complete suppression of gamma-ray absorption. (5) Shift and narrowing of the lines caused by decay interference in the limit of strong driving. (6) Amplification without inversion.

Thus, we conclude that many interesting effects (such as Raman scattering, dynamical Stark splitting, electromagnetically induced transparency, etc.) that have been already widely studied and used in optics should become available in the gamma-ray range. Moreover, completely new effects (such as the shift and narrowing of the lines) appear as a result of the specific symmetry of the compound nuclear-electron system.

These effects provide a number of interesting applications. For example, appearance of new Mössbauer lines shifted by the magnitude of the isomer shift (caused by a change of nuclear radius in the excited state) allows a direct measurement of this shift (and hence a derivation of the relative change of the nuclear radius) that cannot be observed in the traditional Mössbauer spectroscopy. Moreover, laser-Mössbauer spectroscopy permits a measurement of the isomer shift even when it is smaller than the optical linewidth ($\gamma_0 \gg \Delta$) and, hence, cannot be resolved by means of optical spectroscopy.

The hyperfine structure of Mössbauer transitions is caused by an interaction of the nucleus with both the localized electrons (associated with a particular atom) and electrons in the conduction band. Optical driving affords a unique opportunity to study the contribution of a particular electron from the localized outer shell. The method relies on the contribution of this electron to resonant behavior of the optical transition.

Typically, optical transitions in solids are broadened both homogeneously and inhomogeneously. Some situations where homogeneous broadening is narrower than the hyperfine structure have been realized recently in the experiments demonstrating electromagnetically induced transparency in solids [8]. On the other hand, in the limit of a strong driving field ($\Omega \gg \Delta$), neither homogeneous nor inhomogeneous broadening prevents the observation of the Mössbauer spectra modifications.

Prospects for gamma-ray lasers.—Optical driving opens new prospects for gamma-ray lasers. The usual pumping schemes require intense incoherent pump and lead to overheating of the material and destroying the conditions for Mössbauer effect (see a recent exhaustive review [2]). On one hand, this is due to the high resonant losses (which typically exceed nonresonant losses by 3 orders of magnitude). On the other hand, this is caused by the inefficient multistage cascade from the higher-excited energy state to the upper lasing state, since a direct pump at an operating transition in two-level scheme does not provide a population inversion. Coherent optical driving allows both the reduction of resonant losses and the use of direct incoherent pump at the operating transition. The latter may be implemented in two ways: (1) via depletion of the lower lasing level (by means of either optical pumping in a double-Λ scheme or fast decay from this level in a four-level V configuration); (2) via inversionless amplification in V, Λ or double-Λ schemes based on the coherent suppression of resonant absorption.

Reduction of resonant losses and usage of incoherent pumping at the operating transition allow to reduce the required incoherent pump power and heating of an active crystal, by at least 3 orders of magnitude. This transfers the long-standing problem of a gamma-ray laser from the pure speculative theoretical domain into the domain of feasible experiments [9].

Nuclei for experiments.—For observation of most of the effects described above the hyperfine splitting should exceed the Mössbauer linewidth. There are many such materials. Below are some examples.

The interaction between the nuclear spin and the magnetic moment of the ground-state electron of $^{57}Fe^{3+}$ and $^{161}Dy^{3+}$ produces well-resolved hyperfine structure of the order of 8 and 40 mm/s in the Mössbauer transitions [10,11]. The presence of many electronic transitions from the ground state with wavelengths from 0.5 to 1 μm allows for efficient control of the Mössbauer spectra.

The quadrupole splitting of nuclear levels in ^{169}Tm is 79 mm/s [11]. Such a strong quadrupole interaction produces two well-resolved lines at the 8.40 keV Mössbauer transition, in spite of the relatively large natural linewidth of this transition ($\gamma = 9.3$ mm/s). There are several electronic transitions from the ground state with wavelengths between 0.5 and 1 μm that can be used for coherent optical driving.

The Mössbauer measurements for $^{151}Eu^{2+}$ show a chemical isomer shift of 15 mm/s, which is larger than the natural line width [11]. The shift is determined by $\Delta \sim \delta|\psi(0)|^2 \delta R^2/R^2$, where $\delta|\psi(0)|^2$ is the change of the electron density at the nucleus. In these experiments, the change of the electron density was produced by different chemical surroundings of the absorber and emitter. Optical driving of electronic transitions (many of which start from the ground state) provides a different way to vary this density and, hence, to measure the optically induced isomer shift for the same chemical composition.

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