Hyperfine splitting in atoms excited by a resonant laser field

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The behavior of an electron-nucleus system in a resonant laser radiation field is investigated with allowance for the hyperfine interaction. The states of the system are described using the density matrix formalism. The spectrum and the quasienergy level populations are found for the simplest case. Various possible methods for observing the effects under consideration, in particular, using the Mössbauer effect, are discussed.

1. INTRODUCTION

A Mössbauer transition may result in excitation of one of the hyperfine structure (HFS) components of an excited nucleus. Transitions between HFS components may be induced by a high-frequency magnetic field whose frequency is in resonance with the transition frequency between HFS components. This phenomenon is called double NMR-gamma resonance and has been extensively discussed in the literature.[1–4] It has been found experimentally that Rabi splitting of the Mössbauer transition lines occurs in the double resonance effect.[5,6] Rabi splitting is also observed in laser-excited optical electron transitions.[7] A shell electron subjected to the action of laser field and a nucleus may exhibit hyperfine interaction. As a consequence, the electron and the nuclear subsystems are not independent. In this case, the double resonance manifests itself in the fact that one of HFS components is first excited as a result of the Mössbauer transition, for instance, in a resonant absorber, and then the transition to an excited electron level of this atom is induced by the resonant laser field. Such a double resonance can result in repopulation and splitting of HFS components. This process may be realized, for instance, for a doubly charged ion of ^{57}Fe in a paramagnetic host. This case of double resonance is qualitatively different from the double resonance effect for the Zeeman components of the electron subsystem discussed earlier[4,8] in that the electron wave function changes its radial dependence in the process of resonant excitation. The calculation in this case is rather difficult, and so we shall restrict ourselves to the simplest case to clarify the qualitative features of the effect, namely, that in which the nucleus has spin 1/2 and the electron shell can make transitions between the states $S_{1/2}$ and $P_{1/2}$ in consequence of the resonant interaction with the laser. The period of the Rabi oscillations of the valence electron is assumed to be much shorter than the nucleus lifetime for transition to the ground state. The problem is considered for a particular case when only one hyperfine splitting component is initially populated. The analysis of the general case presents no problem.

2. THEORY OF THE EFFECT

Let us consider an atom having two groups of states. In the upper group, the nucleus spin J and the electron spin j^* are equal to 1/2 and are not coupled to each other. The wave function has the form $|JM\rangle \cdot |j^*m^*\rangle$. This group consists of four states, degenerate in energy, denoted by a, b, c, d:

$$a-\left|\frac{1}{2}-\frac{1}{2}\right\rangle\left|\frac{1}{2}-\frac{1}{2}\right\rangle,\quad b-\left|\frac{1}{2}\frac{1}{2}\right\rangle\left|\frac{1}{2}-\frac{1}{2}\right\rangle,$$

$$c-\left|\frac{1}{2}-\frac{1}{2}\right\rangle\left|\frac{1}{2}\frac{1}{2}\right\rangle,\quad d-\left|\frac{1}{2}\frac{1}{2}\right\rangle\left|\frac{1}{2}\frac{1}{2}\right\rangle$$

(see Fig. 1). The lower group of states consists of two hyperfine structure sublevels with a total spin $F=1$ and $F=0$. The wave function is determined by the expression

$$|FM_F\rangle=\sum_{M,m} C^{FM_F}_{JMjm}|JM\rangle\,|jm\rangle, \tag{1}$$

where $C^{FM_F}_{JMjm}$ are the Clebsch–Gordan coefficients relating states of the nuclear spin J and states of the electron shell spin j. This group of levels will be denoted by the numbers 1,2,3 for the states with $F=1$ and the projections $M_F=-1,0,1$, respectively, and 4 for the state with $F=0$.

The laser field can act only on the electron shell. The matrix element for the transition from the state $|j^*m^*\rangle$ to the state $|FM_F\rangle$ under the action of the laser field has the form

$$\langle j^*m^*|\hat{V}|FM_F\rangle=\sum_{M,m} C^{FM_F}_{JMjm}\langle j^*m^*|\hat{V}|jm\rangle. \tag{2}$$

The laser field will be treated as an electromagnetic wave with frequency ω linearly polarized along the z-axis. Then the operator for the interaction between the atomic electron shell and a field of strength E has the form

$$\hat{V}\propto EdY_{10}\cos(\omega t), \tag{3}$$

where d is the dipole moment for a transition between electron states and Y_{10} is the spherical function. By virtue of the Wigner–Eckart theorem, we can write

$$\langle j^*m^*|\hat{V}|jm\rangle\propto C^{j^*m^*}_{jm10}\cos(\omega t). \tag{4}$$

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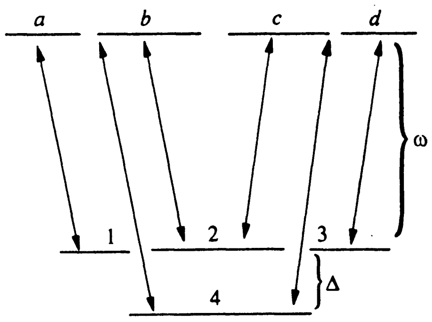

FIG. 1. Level diagram in the system considered and transitions in it under the laser action.

Because of the hyperfine coupling, the electromagnetic field acting on the electron shell alone affects the state of the nucleus, its population and the nuclear transition energies as well.

The time behavior of the system examined is described by the equation for the density matrix $\hat{\rho}$ (Ref. 9):

$$\frac{d\hat{\rho}}{dt}=-\frac{i}{\hbar}[\hat{H}\hat{\rho}], \tag{5}$$

where $\hat{H}$ is the Hamiltonian of the atom in the laser radiation field. In the matrix formulation in the interaction representation using the relations

$$\rho_{mn}=\sigma_{mn}\exp(-i\omega_{mn}t), \quad V_{mn}=(V)_{mn}\exp(-i\omega_{mn}t),$$

where ω_{mn} is the frequency of the transition between the levels m and n, Eq. (5) transforms to an equation for the matrix $\hat{\sigma}$:

$$\frac{d\hat{\sigma}}{dt}=-\frac{i}{\hbar}[\hat{V}\hat{\sigma}]. \tag{6}$$

In the matrix form, neglecting relaxation processes, Eq. (6) is equivalent to the system of equations

$$\dot{\sigma}_{mn}=-\frac{i}{\hbar}\sum_k\{V_{mk}\sigma_{kn}\exp(i\omega_{mk}t)-V_{kn}\sigma_{mk}\exp(i\omega_{kn}t)\}. \tag{7}$$

Equation (7) was derived using the relations $V^*_{mn}=V_{nm}$, $\sigma^*_{mn}=\sigma_{nm}$.

For the level system under consideration, the system of equations (7) breaks up into several independent systems. One of them relates the levels a and 1. It is analogous to the system of equations describing behavior of a two-level system in a radiation field, and we shall not analyze it. A similar system of equations describes the levels d and 3. The levels b, c and 2, 4 are described by a system of equations of order 14, and its solution will be examined in detail.

The laser field will be considered to be in resonance with the levels b, c and levels with $F=1$, i.e., the following relations are valid:

$$\omega=\omega_{b2}, \quad \omega_{b4}=\omega_{b2}+\Delta, \tag{8}$$

where Δ is the hyperfine splitting magnitude.

Introducing new quantities

$$\bar{\sigma}_{b4}=\sigma_{b4}e^{-i\Delta t}, \quad \bar{\sigma}_{24}=\sigma_{24}e^{-i\Delta t}, \quad \bar{\sigma}_{c4}=\sigma_{c4}e^{-i\Delta t},$$

the system of equations (7) can be recast as a system of differential equations with constant coefficients:

$$\begin{aligned}
&\dot{\sigma}_{b2}=-iW(\sigma_{bb}-\sigma_{22})+iW\bar{\sigma}_{42},\\
&\dot{\sigma}_{c2}=iW(\sigma_{cc}-\sigma_{22})+iW\bar{\sigma}_{42},\\
&\dot{\sigma}_{b4}+i\Delta\bar{\sigma}_{b4}=-iW(\sigma_{bb}-\sigma_{44})+iW\bar{\sigma}_{24},\\
&\dot{\sigma}_{c4}+i\Delta\bar{\sigma}_{c4}=-iW(\sigma_{cc}-\sigma_{44})-iW\bar{\sigma}_{24},\\
&\dot{\sigma}_{22}=iW(\sigma_{b2}-\sigma_{2b})-iW(\sigma_{c2}-\sigma_{2c}),\\
&\dot{\sigma}_{44}=-iW(\bar{\sigma}_{4c}+\bar{\sigma}_{4b})+iW(\bar{\sigma}_{b4}+\bar{\sigma}_{c4}),\\
&\dot{\sigma}_{24}+i\Delta\bar{\sigma}_{24}=-iW(-\bar{\sigma}_{b4}+\bar{\sigma}_{c4}+\sigma_{2b}+\sigma_{2c}),\\
&\dot{\sigma}_{bb}=iW(\sigma_{2b}-\sigma_{b2})+iW(\bar{\sigma}_{4b}-\bar{\sigma}_{b4}),\\
&\dot{\sigma}_{cc}=-iW(\sigma_{2c}-\sigma_{c2})+iW(\bar{\sigma}_{4c}-\bar{\sigma}_{c4}).
\end{aligned} \tag{9}$$

For σ_{2b}, σ_{2c}, $\bar{\sigma}_{4b}$, $\bar{\sigma}_{4c}$, and $\bar{\sigma}_{42}$, the equations have the complex-conjugate form. Here, $W=Ed/2\hbar\sqrt{6}$. Performing transformations, we can get an equation for σ_{22} from (9):

$$\ddot{\sigma}_{22}=2W^2(\sigma_{bb}+\sigma_{cc}-2\sigma_{22}). \tag{10}$$

Let us introduce the combinations $\sigma_+=\bar{\sigma}_{4b}+\bar{\sigma}_{4c}$, $\sigma^*_+=\bar{\sigma}_{b4}+\bar{\sigma}_{c4}$. According to (9), the following relations hold for σ_+ and σ^*_+:

$$\begin{aligned}
&\dot{\sigma}_+-i\Delta\sigma_+=iW(\sigma_{bb}+\sigma_{cc}-2\sigma_{44}),\\
&\dot{\sigma}^*_++i\Delta\sigma^*_+=-iW(\sigma_{bb}+\sigma_{cc}-2\sigma_{44}).
\end{aligned} \tag{11}$$

It follows that

$$\begin{aligned}
&i\Delta(\sigma^*_+-\sigma_+)+\dot{\sigma}^*_++\dot{\sigma}_+=0,\\
&i\Delta(\sigma^*_++\sigma_+)+\dot{\sigma}^*_+-\dot{\sigma}_+=-2iW(\sigma_{bb}+\sigma_{cc}-2\sigma_{44}).
\end{aligned} \tag{12}$$

From (9), the relation

$$i\dot{\sigma}_{44}=W(\sigma_+-\sigma^*_+) \tag{13}$$

is valid for σ_{44}. Using the second equation of (12) and (13), we get for σ_{44}

$$\frac{i}{W}\ddot{\sigma}_{44}=i\Delta(\sigma^*_++\sigma_+)+2iW(\sigma_{bb}+\sigma_{cc}-2\sigma_{44}). \tag{14}$$

From the first equation of (12) and from (13), a relation for the first derivatives follows:

$$\frac{\Delta}{W}\dot{\sigma}_{44}=-(\dot{\sigma}^*_++\dot{\sigma}_+). \tag{15}$$

Integrating (15) under the assumption that at $t=0$ the population of the level 4 is equal to 1, we find

$$\sigma^*_++\sigma_+=-\frac{\Delta}{W}(\sigma_{44}-1). \tag{16}$$

With allowance for (16) and the normalization condition

$$\sigma_{bb}+\sigma_{cc}+\sigma_{22}+\sigma_{44}=1, \tag{17}$$

Eq. (14) is transformed into

$$\ddot{\sigma}_{44}+\sigma_{44}(\Delta^2+6W^2)+2W^2\sigma_{22}-\Delta^2-2W^2=0. \quad (18)$$

In view of the normalization condition (17), the equation for σ_{22} will appear as follows:

$$\ddot{\sigma}_{22}+6W^2\sigma_{22}+2W^2\sigma_{44}-2W^2=0. \quad (19)$$

Equations (18) and (19) comprise a closed system of linear inhomogeneous differential equations with constant coefficients. The characteristic equation corresponding to a system of homogeneous equations determines the state energies of the quantum system examined interacting with the resonance field:

$$k=\pm\sqrt{-\mu\pm\sqrt{\nu}}, \quad \mu=6W^2+\frac{\Delta^2}{2}, \quad \nu=4W^4+\frac{\Delta^4}{4}. \quad (20)$$

Note that we have $\mu > \sqrt{\nu}$ and consequently k has only imaginary values. The solution is sought in the form

$$\sigma_j=A_j+\sum_{i=1}^{4}\alpha_i^{(j)}\exp(k_i t), \quad (21)$$

where k_i are defined in (20), and the constants A_j and $\alpha_i^{(j)}$ are determined from the inhomogeneous system of equations and initial conditions. Finally, we get for the initial conditions $\sigma_{44}=1$, $\sigma_{22}=0$, $\sigma_{bb}=0$, $\sigma_{cc}=0$:

$$\sigma_{22}=\left\{4W^2+\left(\frac{\Delta^2}{2}+\sqrt{\nu}\right)\left[3\left(\frac{\Delta^2}{2}-\sqrt{\nu}\right)\right.\right.$$
$$\left.-2W^2\right]\frac{\cos(\sqrt{\mu-\sqrt{\nu}}t)}{\sqrt{\nu}}-\left(\frac{\Delta^2}{2}-\sqrt{\nu}\right)\left[3\left(\frac{\Delta^2}{2}+\sqrt{\nu}\right)\right.$$
$$\left.\left.-2W^2\right]\frac{\cos(\sqrt{\mu+\sqrt{\nu}}t)}{\sqrt{\nu}}\right\}\frac{1}{(3\Delta^2+16W^2)},$$

(22)

$$\sigma_{44}=\left\{3\Delta^2+4W^2-2W^2\left[3\left(\frac{\Delta^2}{2}-\sqrt{\nu}\right)\right.\right.$$
$$\left.-2W^2\right]\frac{\cos(\sqrt{\mu-\sqrt{\nu}}t)}{\sqrt{\nu}}+2W^2\left[3\left(\frac{\Delta^2}{2}+\sqrt{\nu}\right)\right.$$
$$\left.\left.-2W^2\right]\frac{\cos(\sqrt{\mu+\sqrt{\nu}}t)}{\sqrt{\nu}}\right\}\frac{1}{(3\Delta^2+16W^2)}.$$

As seen from (22), the values of k_i from (20) determine the oscillation frequencies of the level populations. In a strong field ($W\gg\Delta$) the time-average populations of the levels 2 and 4 are equal to 1/4. In a weak field, the population of level 2 will be proportional to $(4/3)(W/\Delta)^2$. The Rabi oscillation frequencies are shown in Fig. 2 as functions of the parameter W/Δ. The solid curve corresponds to $X = \sqrt{\mu+\sqrt{\nu}}/\Delta$ and the dashed curve corresponds to $Y = \sqrt{\mu-\sqrt{\nu}}/\Delta$.

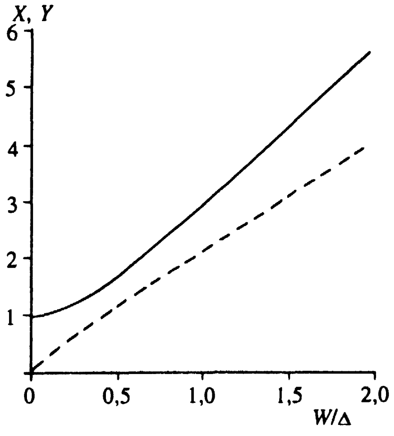

FIG. 2. Rabi frequencies as functions of laser power.

3. DISCUSSION AND CONCLUSION

Let us discuss various possible methods for observing the effects considered above. Assume that a Mössbauer transition occurs from a system of electron-nucleus states. Since the values of the squares of electron wave function in the states (1–4) and (*a*–*d*) are different, the action of a strong laser field on an atom of the Mössbauer source will result in the appearance, along with the old nuclear transition line, of a new line shifted by an amount proportional to the square of the difference of the electron wave functions. Using a resonant absorber, one can observe variations of the Mössbauer absorption spectrum when the absorber is acted upon by the laser field. With the laser power growing, a nuclear transition line from the hyperfine splitting component, which is not populated by the incident Mössbauer radiation, should appear in the spectrum. In a weak laser field, the intensity of this line will be proportional to $(W/\Delta)^2$, according to (22). From this intensity, the hyperfine splitting magnitude can be estimated. For a strong laser field, splitting of Mössbauer spectrum lines should be observed in accordance with (20). Finally, one can detect modulation of the Mössbauer transition intensity with the Rabi frequency according to Eqs. (22).

To observe Rabi splitting, the period of Rabi oscillations should be less than the Mössbauer level lifetime which is usually no smaller than 10^{-6}–10^{-8} s. Moreover, this period should be less than the electron shell relaxation time. The latter criterion proves to be the most stringent, because the transverse relaxation time of an electron shell is 10^{-11}–10^{-13} s (Ref. 9). Therefore, to excite atoms in the Rabi mode, low temperatures and pulsed tunable lasers are needed. It is assumed that the longitudinal relaxation time of an electron shell is greater than the nuclear decay period. The field strength in the laser pulse should be at least 10^4 V/cm. Apparently, copper vapor lasers are the most suitable for studying the effect under consideration. They possess high pulse repetition rate and high peak power.

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