Resonant excitation of hyperfine levels in a bichromatic laser wave in connection with nuclear decay

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A new nuclear-spectroscopy method is proposed. It is based on a resonant interaction of an atom which has hyperfine structure in its ground state with the field of a bichromatic laser wave. The method is essentially free of Doppler-broadening effects. The model of a Λ system is used to derive expressions for the time evolution of the populations of the hyperfine components and for the behavior of the angular polarization of the emission from the nuclear decay as a function of the relations between the strengths and phases of the fields in the bichromatic wave. Ways to implement this method experimentally are proposed. © *1994 American Institute of Physics.*

A new method of nonlinear spectroscopy, based on the coherent trapping of populations in three-level systems, has recently won recognition.[1,2] This effect has also been observed in ^{23}Na atoms, which have a hyperfine structure.[3] In stable atoms, the effect is manifested by a lowering of the level of spontaneous emission. In unstable atoms, the spontaneous intensity may be very small, and one might prefer to observe nuclear polarization by studying the angular distribution of nuclear emission. The method of optical nuclear polarization in a circularly polarized laser field[4] (RADOP) runs into difficulties when applied to gases, because of the Doppler effect.

In this letter we discuss a resonant interaction of a bichromatic laser field with an atom which has hyperfine structure in its ground state. In this case, Doppler broadening has essentially no effect. In addition, the atomic and nuclear polarization can be achieved over a time shorter than the relaxation time of the electron cloud.

We consider a plane-polarized bichromatic laser wave with frequencies ω_1 and ω_2 ($\omega_2 - \omega_1 = \delta\omega \ll \omega_{1,2}$) and a three-level Λ atom. We assume that both components of the laser wave are at resonance with atomic transitions between corresponding levels of the hyperfine structure (we will call these levels 1 and 2) and state 3, in which the electron cloud is excited. We assume that there is no interaction between nucleus and the electrons of the cloud. We denote the wave functions of these states by $\Psi_i(t)$. We assume that the relaxation time of level 3 is much longer than the nuclear lifetime. Under these conditions we can use the wave-function method to describe the quantum system, and we can ignore relaxation of the electron cloud.

The equations of time-dependent perturbation theory for the amplitudes of the states of this system are written as follows in the resonant approximation:

$$\frac{da_1(t)}{dt} = -iV_1 e^{i\varphi_1} a_3(t),$$

$$\frac{da_2(t)}{dt} = -iV_2 e^{i\varphi_2} a_3(t),$$

$$\frac{da_3(t)}{dt} = -iV_1 e^{-i\varphi_1} a_1(t) - iV_2 e^{-i\varphi_2} a_2(t), \tag{1}$$

where $V_{1,2}$ are the dipole matrix elements for the transition caused between the corresponding hyperfine level and level 3 by the field of the frequency $\omega_{1,2}$, which has the phase $\varphi_{1,2}$ and which is at resonance with this transaction. The solution of Eqs. (1) for the initial conditions $a_1(0)=A_1$, $a_2(0)=A_2$, $a_3(0)=0$ is

$$a_1(t)[A_1V_2^2 - A_2V_1V_2e^{-i\xi} + (A_1V_1^2 + A_2V_1V_2e^{-i\xi})\cos(\Omega t)]/\Omega^2,$$

$$a_2(t) = [A_2V_1^2 - A_1V_1V_2e^{i\xi} + (A_2V_2^2 + A_1V_1V_2e^{i\xi})\cos(\Omega t)]/\Omega^2,$$

$$a_3(t) = -i[A_1V_1e^{-i\varphi_1} + A_2V_2e^{-i\varphi_2}]\sin(\Omega t)/\Omega. \tag{2}$$

Here $\xi = \varphi_2 - \varphi_1$ and $\Omega^2 = V_1^2 + V_2^2$. It can be seen from (2) that the population of level 3 may be zero at an arbitrary time if the phases and strengths of the laser fields satisfy certain relations. This "coherent trapping of populations" occurs under the condition

$$\cos\xi = -\frac{A_1^2V_1^2 + A_2^2V_2^2}{2A_1A_2V_1V_2}. \tag{3}$$

We have the following expression for the time-average difference between the populations of the hyperfine levels (the nuclear polarization):

$$\overline{|a_2(t)|^2} - \overline{|a_1(t)|^2} = (V_2^2 - V_1^2)[A_1^2(V_1^2/2 - V_2^2) + A_2^2(V_2^2/2 - V_1^2) + 3A_1A_2V_1V_2\cos\xi]. \tag{4}$$

The superior bar means a time average. In the case of coherent population trapping this difference is zero.

It is interesting to analyze how a bichromatic field would affect nuclear decay in a transition from this system of electron–nuclear levels to a final state $\Psi_0(t)$ in which there is no hyperfine splitting or resonant interaction of the electron cloud with the laser field, and the nucleus is in its ground state. The spectrum of the nuclear emission can be calculated by perturbation theory, by calculating the square modulus of the time integral of a matrix element of the nuclear-transition operator $\hat{W}_\gamma$:

$$S = \int_0^\infty dt \left\langle \sum_{i=1}^{3} a_i(t)\Psi_i(t)|\hat{W}_\gamma|\Psi_0(t) \right\rangle. \tag{5}$$

Carrying out the integration, we find

$$S = \sum_i D_i(\vartheta,\varphi)[\beta_{i1}\delta(\omega_i) + \beta_{i2}\delta(\omega_i + \Omega) + \beta_{i3}\delta(\omega_i - \Omega)]. \tag{6}$$

Here $\delta(\omega)$ is the Dirac δ-function of ω; $\omega_i = E_\gamma - E_i + E_0$, where E_γ is the energy of the γ ray, E_i is the energy of atomic state i, and E_0 is the energy of the atom in state Ψ_0; and $D_i(\vartheta,\varphi)$ are the matrix elements of the nuclear emission from the state Ψ_i in the direc-

tion specified by the angles (ϑ,φ). Since the coefficients $\beta_{i,j}$ depend on the parameters of the laser fields, the intensities of the individual lines in the spectrum of the nuclear emission depend on these parameters.

Evaluating the integral $W\sim\int dE_\gamma|S|^2$, we find the following expression for the angular anisotropy of the nuclear emission:

$$X(\vartheta,\varphi)=[(A_1^2V_2^2+A_2^2V_1^2-2A_1A_2V_1V_2\cos\xi)(D_1^2V_2^2+D_2^2V_1^2)/\Omega^4$$
$$+(A_1^2V_1^2+A_2^2V_2^2+2A_1A_2V_1V_2\cos\,\xi)(D_1^2V_1^2+D_2^2V_2^2$$
$$+D_3^2V_1^2+D_3^2V_2^2)/2/\Omega^4-D_1^2-D_2^2]/(D_1^2+D_2^2). \quad (7)$$

It follows from (7) that under conditions of coherent population trapping the angular distribution of γ rays is isotropic.

If the interaction of the atom with the field is strong enough that the Rabi frequency is greater than the longitudinal relaxation width, the atomic levels split by an amount equal to the Rabi frequency (this is Rabi splitting).[5,6] For components of the hyperfine structure and for a resonant bichromatic field, this splitting occurs for both components of the hyperfine structure. At a certain relation between the intensities of the fields of the bichromatic wave, the components of the Rabi splitting of both hyperfine levels may cross. This crossing may be manifested in a change in the angular distribution of the products of the nuclear decay. In the expression for W, this crossing is manifested by the appearance of additional cross terms in the case in which the arguments of the δ -functions are the same. For the relation $\Omega=\Delta$, for example, the additional terms in W are

$$\tilde{W}\sim D_1D_2\{A_1A_2[V_1^4+V_2^4-2V_1^2V_2^2\cos(2\xi)]+(A_1^2-A_2^2)V_1V_2(V_2^2-V_1^2)\cos\xi\}/\Omega^4. \quad (8)$$

Under the condition $\Omega=\Delta/2$, we have

$$\tilde{W}\sim D_1D_2V_1V_2\cos\,\xi[A_1^2V_1^2+A_2^2V_2^2+2A_1A_2V_1V_2\cos\,\xi]/\Omega^4/2$$
$$+D_1D_3[(A_1^2-A_2^2)V_1V_2^2\cos\,\varphi_1+A_1A_2V_2^3\cos\,\varphi_2-A_1A_2V_2V_1^2\cos(\xi-\varphi_1)]/\Omega^3$$
$$+D_2D_3[(A_1^2-A_2^2)V_2V_1^2\cos\,\varphi_2+A_1A_2V_1V_2^2\cos(\xi+\varphi_2)-A_1A_2V_1^3\cos\,\varphi_1]/\Omega^3. \quad (9)$$

In the case $\Omega=\Delta/4$, on the other hand, we have

$$\tilde{W}\sim[A_1^2V_1^2+A_2^2V_2^2+2A_1A_2V_1V_2\cos\,\xi](D_1D_3V_1\cos\,\varphi_1-D_2D_3V_2\cos\,\varphi_2)/\Omega^3/2. \quad (10)$$

The onset of these terms gives rise to an additional anisotropy of the products of the nuclear decay, which depends on the strengths of the laser fields.

We believe that these effects may find broad applications, and that various methods could be used to observe them. For example, to measure hyperfine splitting one could tune the laser fields to resonance with one of the transitions in the system of atomic levels and vary the frequency of the other laser. The interaction of the hyperfine system of atomic levels with a monochromatic resonant laser field and also the effect of this interaction on nuclear decay were studied in Ref. 6. When the frequency of the second laser reaches resonance with the second transition, the angular distribution of the nuclear emission becomes anisotropic. When the strengths of the laser fields are equal, under

conditions that their phase difference is a multiple of π [see (3) with $A_1=A_2$], a coherent trapping of population occurs, and the angular anisotropy should disappear again. The line of nuclear emission from a state with an excited cloud, which arises (as was shown in Ref. 6) from an interaction with a resonant electromagnetic field, also disappears. In this case the difference between the frequencies of the components of the bichromatic wave will correspond to the magnitude of the hyperfine splitting.

In another approach we suggest using as the Λ system the Zeeman components of hyperfine levels which are interacting with the two laser waves. By varying the strength of the magnetic field at a fixed difference between the laser fields ($\delta\omega \ll \Delta$), one should be able to reach resonance between the system of Zeeman components and the bichromatic field. An experiment of this type might make it possible to measure the g-factors of unstable nuclear states.

We wish to stress that these measurements would be carried out under conditions such that the Doppler effect has essentially no influence. In an atom moving at a velocity $\mathbf{v}$, a resonance at each of the frequencies occurs under the condition $\omega_\nu + \mathbf{k}\cdot\mathbf{v} = \omega_{\nu,3}$, where ω_ν is the frequency of field ν ($\nu=1,2$). Consequently, the coherent population trapping occurs under the condition $\omega_1 - \omega_2 + \mathbf{k}_1\cdot\mathbf{v}_1 - \mathbf{k}_2\cdot\mathbf{v}_2 \approx \Delta$ in our case. The difference between the frequencies of the bichromatic wave may thus differ from the resonant values by an amount on the order of $\Delta v/c$. Even if the Doppler width of the resonant transitions, which is $\approx \omega_\nu v/c$, is far greater than the hyperfine splitting Δ, one could observe the coherent population trapping by the method of varying the difference between the frequencies of lasers of identical power and thereby determine measurable quantities, e.g., the hyperfine splitting within an error $\Delta v/c \ll \omega_\nu v/c$. These effects could thus be observed in gases, and ultralow temperatures would not be required for experiments on solids.

When the method proposed here is used, the polarization of the nucleus may arise over a time far shorter than the spontaneous relaxation time. This method, in contrast with RADOP, may thus find applications in measurements of nuclear parameters in cases in which the nuclear lifetime is shorter than the spontaneous-relaxation time of the electron cloud.

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