RADIOOPTICAL RESONANCE OF ATOMS IN STRONG MAGNETIC FIELDS

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Submitted to JETP editor November 11, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) **49**, 163-169 (July, 1965)

Resonance phenomena are considered in a system possessing an electron angular momentum J and nuclear moment I in a strong magnetic field. An expression is derived for the intensity of reradiated light when microwave and radio-frequency fields are applied to the system. It is shown that the light intensity changes appreciably on approach to the nuclear resonance frequency. This phenomenon can be employed for investigating experimentally the hyperfine structure of atoms.

THE proposal of Brossel and Bitter[1] to use double radiooptical resonance with atoms in an excited state to study experimentally the energy structure has led to the discovery of a new phenomenon[2] referred to in scientific literature as light beats. The essence of this phenomenon is as follows. The light emitted by atoms during a transition from various sublevels of an excited state to the ground state is incoherent. However, if the atoms are subjected to the influence of an oscillating magnetic field which induces magnetic dipole transitions between the excited sublevels, then the state of atoms originally excited in one of the eigenstates becomes a coherent superposition of all those states in which the atoms can exist under radio-frequency transitions. This coherence is manifest in the fluorescent radiation as interference between different frequencies.

In the case of a strong magnetic field, it is possible to separate the terms in the Hamiltonian which describe the electronic and nuclear systems only. This allows the possibility of carrying out electron-nuclear double resonance in an excited state. Resonance with electrons and nuclei affects differently the properties of the light emitted by the atom during the transition from an excited state to the ground state. The microwave field inducing magnetic dipole transitions between the electron sublevels, as well as the radio-frequency field which causes transitions between the magnetic sublevels of the nucleus, have a definite phase relationship with the vibrational phases of the electron and nuclear sublevels. This establishes definite phase relationship between the various states. Unlike the case of the electron system, the coherence created in the nuclear system does not cause emitted-light intensity beats at the frequency of the radio-frequency field. However, in this case the amplitude of the emitted modulated light, as well as the time-constant intensity component, depend on the resonance conditions within the nuclear system. When there is no radio-frequency field, the intensity of the reradiated light is described by a well known expression[2].

1. EVOLUTION OF THE SYSTEM'S STATE IN TIME

If an atom is placed in a strong magnetic field $\mathbf{H}_0$, the interaction of its electron angular momentum $\mathbf{J}$ with the nuclear moment $\mathbf{I}$ can be neglected by comparison with the interaction with the field $\mathbf{H}_0$. In this case the electron and nuclear moments precess independently about the direction of the magnetic field. The Hamiltonian of an atomic system in a magnetic field $\mathbf{H}_0$ interacting with a microwave field $\mathbf{h}^J$, a radio-frequency field $\mathbf{h}^I$, and light fields can be written as

$$\mathcal{H} = \mathcal{H}_0 + \sum_{\mu=0,\pm1} \gamma_J J_\mu h_{-\mu}{}^J + \sum_{\mu=0,\pm1} \gamma_I I_\mu h_{-\mu}{}^I + \mathcal{H}_{opt} + \mathcal{H}_\Gamma, \tag{1}$$

where

$$J_{\pm1} = 2^{-1/2}(J_x \pm iJ_y), \quad I_{\pm1} = 2^{-1/2}(I_x \pm iI_y),$$

$$J_0 = J_z, \quad I_0 = I_z, \quad h_{\pm1}{}^J = 2^{-1/2} h^J e^{\pm i\omega_2 t},$$

$$h_{\pm1}{}^I = 2^{-1/2} h^I e^{\pm i\omega_1 t}, \quad h_0{}^I = h_0{}^J = H_0. \tag{2}$$

The second and third terms in (1) describe the interaction of the atom with a constant and with oscillating fields. The fourth term describes the interaction of the j-th atom in question with the radiation field:

$$\mathcal{H}_{opt} = E_j(t)\mathbf{e}_j{}^0\mathbf{D} = E_j(t)P, \tag{3}$$

where D is the electric dipole moment operator, and e_j^0 is the unit vector of the light-wave polarization. The last term $\mathscr{H}_\Gamma$ describes the radiation damping of the excited state. For the sake of simplicity, let us assume that the decay rate for all the sublevels of the excited state is identical and given by the constant Γ.

The time variation of the state $|t\rangle$ satisfies Schrödinger's equation:

$$i\hbar\frac{d}{dt}|t\rangle = \mathscr{H}|t\rangle. \qquad (4)$$

We shall make no further assumptions concerning the smallness of the amplitudes of the microwave and radio-frequency fields, which may be large enough to saturate the magnetic transitions. However, the interaction of the atom with the radiation field will be computed, as in [3], accurate to first order of perturbation theory.

Let us assume that the atomic state can be represented at any instant of time as

$$|t\rangle = a_{m_0n_0}(t)\,|m_0n_0\rangle + \sum_{m,\,n\neq m_0,\,n_0} a_{mn}(t)\,|mn\rangle, \qquad (5)$$

where m_0 and n_0 are quantum numbers for the electrons and nucleus in the ground state and m and n specify the energy levels of the atom in an excited state.

Let us put (1) in the form $\mathscr{H} = \mathscr{H}' + \mathscr{H}_{opt}$, where

$$\mathscr{H}' = \mathscr{H}_0 + \mathscr{H}_\Gamma + \sum_{\mu=0,\,\pm1}\gamma_J J_\mu h_{-\mu}^J + \sum_{\mu=0,\,\pm1}\gamma_I I_\mu h_{-\mu}^I. \qquad (6)$$

Let $|\rangle$ denote the state of the system at time t, which has evolved from $|t_0\rangle$ only under the influence of $\mathscr{H}'$. The corresponding equation of motion takes the form

$$i\hbar\frac{d}{dt}|\rangle = \mathscr{H}'|\rangle. \qquad (7)$$

The transformation from $|t_0\rangle$ to $|\rangle$ can be expressed by using the operator $U(t, t_0)$ in the following manner[3]:

$$U(t, t_0)\,|t_0\rangle = |\rangle. \qquad (8)$$

We then obtain the solution to (4) in the form

$$|t\rangle = U(t, 0)\,|m_0n_0\rangle + \frac{1}{i\hbar}\int_0^t U(t, t_0)\mathscr{H}_{opt}(t_0)U(t_0, 0)\,dt_0|m_0n_0\rangle. \qquad (9)$$

In order to find $U(t, t_0)$, and, consequently, the coefficients a_{mn} in equation (5) as well, we carry out a unitary transformation:

$$|\rangle' = T|\rangle = \exp(i\omega_1 I_z t/\hbar)\exp(i\omega_2 J_z t/\hbar). \qquad (10)$$

Inserting this in (7), we obtain

$$i\hbar\frac{d}{dt}|\rangle' = (\mathscr{H}_0 + \mathscr{H}_\Gamma + \Omega_I I_x + \delta_I I_z + \Omega_J J_x + \delta_J J_z)\,|\rangle' = \overline{\mathscr{H}}|\rangle', \qquad (11)$$

where we have introduced the notation

$$\Omega_I = \gamma_I h^I, \quad \Omega_J = \gamma_J h^J;$$

$$\delta_I = \gamma_I H_0 - \omega_1 = \omega_I - \omega_1, \quad \delta_J = \gamma_J H_0 - \omega_2 = \omega_J - \omega_2.$$

The solution to Eq. (11) is easily found:

$$|\rangle' = \exp\{\widetilde{\mathscr{H}}(t - t_0)/i\hbar\}\,|t_0\rangle'. \qquad (12)$$

Applying the inverse transformation T^{-1} to (12), we obtain

$$|\rangle = \exp\{-i(\omega_1 I_z + \omega_2 J_z)t/\hbar\}\exp\{\widetilde{\mathscr{H}}(t - t_0)/i\hbar\} \times \exp\{i(\omega_1 I_z + \omega_2 J_z)t_0/\hbar\}\,|t_0\rangle. \qquad (13)$$

From this we derive an expression for $U(t, t_0)$

$$U(t, t_0) = \exp\{-i(\omega_1 I_z + \omega_2 J_z)t/\hbar\} \times \exp\{\widetilde{\mathscr{H}}(t - t_0)/i\hbar\}\exp\{i(\omega_1 I_z + \omega_2 J_z)t_0/\hbar\}. \qquad (14)$$

In order to find the coefficients a_{mn} we take a product of the form

$$\langle mn|t\rangle = a_{mn} = \frac{1}{i\hbar}\int_0^t dt_0\,\langle mn|U(t, t_0)\mathscr{H}_{opt}(t_0)U(t_0, 0)\,|m_0n_0\rangle$$

$$= \frac{1}{i\hbar}\int_0^t dt_0 \sum_{m'n'm''n''}\langle mn|U(t, t_0)\,|m'n'\rangle \times \langle m'n'|\mathscr{H}_{opt}(t_0)\,|m''n''\rangle \times \langle m''n''|U(t_0, 0)\,|m_0n_0\rangle. \qquad (15)$$

The terms on the right side of (15) are in the form of products which can be computed without difficulty.

Let us find the matrix elements of the operator $U(t, t_0)$:

$$\langle mn|U(t, t_0)\,|m'n'\rangle = \exp\{-i(\omega_1 n + \omega_2 m)t\}\exp\{i(\omega_1 n' + \omega_2 m')t_0\} \times \exp\{-i(k_0 - i\Gamma/2)(t - t_0)\}$$
$$\times \langle m|\exp\{\widetilde{\mathscr{H}}_J(t - t_0)/i\hbar\}\,|m'\rangle \times \langle n|\exp\{\widetilde{\mathscr{H}}_I(t - t_0)/i\hbar\}\,|n'\rangle,$$

$$\widetilde{\mathscr{H}}_J = \Omega_J J_x + \delta_J J_z, \quad \widetilde{\mathscr{H}}_I = \Omega_I I_x + \delta_I I_z. \qquad (16)$$

Equation (16) is valid because the electron system is independent of the nuclear system. In the case of an arbitrary field this form cannot be used and the calculation of the matrix elements $U(t, t_0)$ becomes very complicated. This question will be examined in greater detail elsewhere.

There now remains the task of computing the matrix elements which appear in (16). The matrix elements $\langle m | \exp\{\widetilde{\mathscr{H}}_J(t - t_0)/i\hbar\} m' \rangle$ are most simply found in a new coordinate system, rotated with respect to the original such that the axis oz' of the new system coincides with the direction of the effective magnetic field H^J_{eff}:

$$H^J_{eff} = (\delta_J^2 + \Omega_J^2)^{1/2} \gamma_J^{-1}. \qquad (17)$$

Obviously the coordinate system to be used for the nuclear spins of such a system is that in which the oz'' axis coincides with the direction of the effective field H^I_{eff}:

$$H^I_{eff} = (\delta_I^2 + \Omega_I^2)^{1/2} \gamma_I^{-1}. \qquad (18)$$

The state vectors are transformed with the help of the expression

$$|m\rangle = \sum_\mu |\mu\rangle \langle \mu | m\rangle. \qquad (19)$$

Taking into consideration (17), (18), and (19), we obtain for $\langle mn | U(t_0, t) | m'n' \rangle$

$$\langle mn | U(t, t_0) | m'n' \rangle = \exp\{-i(\omega_1 n + \omega_2 m) t\} \exp\{i(\omega_1 n' + \omega_2 m') t_0\} \times \sum_{\mu\zeta} \langle m | \mu\rangle\langle \mu | m'\rangle\langle n | \zeta\rangle\langle \zeta | n'\rangle \times \exp\{-i\lambda_\mu (t - t_0)\} \exp\{-i\lambda_\zeta (t - t_0)\}; \qquad (20)$$

$$\hbar\lambda_\mu = \hbar(k_0 - i\Gamma/2 + \mu\gamma_J H^J_{eff}), \qquad \hbar\lambda_\zeta = \hbar\zeta H^I_{eff}\gamma_I. \qquad (21)$$

The matrix elements $\langle m''n'' | U(t, 0) | m_0 n_0 \rangle$, which refer to the sublevels of the ground state, are computed in a similar fashion:

$$\langle m''n'' | U(t_0, 0) | m_0 n_0 \rangle = \exp\{-i(\omega_1 n_0 + \omega_2 m_0) t_0\} \times \exp\{-i(\omega_{J'} - \omega_2) m_0 t_0\} \exp\{-i(\omega_{I'} - \omega_1) n_0 t_0\}. \qquad (22)$$

Here we have utilized the fact that the frequencies ω_1 and ω_2 are far from the resonance frequencies of the atoms in the ground state. Now we obtain for the state $|t\rangle$ the following expression:

$$|t\rangle = a_{m_0 n_0} | m_0 n_0\rangle + \frac{1}{i\hbar} \int_0^t dt_0 E_j(t_0) \sum_{mnm'n'\mu\zeta} \exp\{-i(\omega_1 n + \omega_2 m) t\} \times \exp\{i(\omega_1 n' + \omega_2 m') t\} \exp\{-i\lambda_\mu (t - t_0)\} \times \exp\{-i\lambda_\zeta (t - t_0)\} \exp\{-i m_0 \omega_{J'} t_0\} \exp\{-i n_0 \omega_{I'} t_0\} \times \langle m | \mu\rangle\langle \mu | m'\rangle\langle n | \zeta\rangle\langle \zeta | n'\rangle P_{m'n'm_0n_0} | mn\rangle,$$

$$P_{m'n'm_0n_0} = \langle m'n' | P | m_0 n_0\rangle. \qquad (23)$$

2. CALCULATION OF THE INTENSITY OF FLUORESCENT RADIATION

The energy flux radiated by an atomic system in some direction denoted by the unit vector ϵ is given by the expression

$$\mathbf{S} = \frac{c}{8\pi} (\mathbf{E}^*\mathbf{E}) \boldsymbol{\varepsilon}. \qquad (24)$$

The component of the radiation field in the direction $\mathbf{e}^0$ lying in a plane normal to ϵ is equal to[4]

$$\mathbf{e}^0\mathbf{E} = \frac{2k^2}{rc^2} \langle m_0 n_0 | \mathbf{e}^0 \mathbf{D} | t\rangle. \qquad (25)$$

Thus the expression for the intensity S depends upon the form of the matrix elements $\langle m_0 n_0 | \mathbf{e}^0 \cdot \mathbf{D} | t\rangle$:

$$\langle m_0 n_0 | \mathbf{e}^0 \mathbf{D} | t\rangle = \frac{1}{i\hbar} \int_0^t dt_0 E_j(t_0) \sum_{mnm'n'\zeta\mu} G_{m_0 m} \langle m | \mu\rangle\langle \mu | m'\rangle \times P_{m'm_0} \langle n_0 | \zeta\rangle\langle \zeta | n_0\rangle \exp\{-i(\omega_1 n_0 + \omega_2 m) t\} \times \exp\{-i\lambda_\zeta (t - t_0)\} \exp\{-i\lambda_\mu (t - t_0)\} \times \exp\{i(\omega_1 n_0 + \omega_2 m') t_0\} \times \exp\{-i(m_0 \omega_{J'} + n_0 \omega_{I'}) t_0\}. \qquad (26)$$

Using (26) and (24), we obtain the following expression for the intensity of light linearly polarized in the direction $\mathbf{e}^0$ at time t[3]:

$$S = \frac{k_0^4 \rho(k_0)}{2\pi c^3 \hbar^2 r_0^2} \sum_{\substack{mm'll' \\ \mu\mu'\zeta\zeta'}} P_{m'l'} \langle m | \mu\rangle\langle \mu | m'\rangle\langle l' | \mu'\rangle\langle \mu' | l\rangle \times C_{ml} \langle n_0 | \zeta\rangle\langle \zeta | n_0\rangle\langle n_0 | \zeta'\rangle\langle \zeta' | n_0\rangle \times \{\Gamma - i[(\mu' - \mu)\gamma_J H^J_{eff} + (\zeta' - \zeta)\gamma_I H^I_{eff}] + \omega_2(l' - m')\}^{-1} \exp\{i\omega_2(l - l' + m' - m) t\}, \qquad (27)$$

where $\rho(k_0)$ is the spectral density of the incident radiation. In the derivation of (27) we introduced the abbreviated notation

$$P_{m'l'} = P_{mm_0} P_{m_0 l'}, \qquad C_{ml} = G^*_{mm_0} G_{m_0 l}.$$

The indices m, m', l, l', μ, and μ' can assume the values $-J, -J + 1, \ldots, J - 1, J$; the indices ζ and ζ' can assume the values $-I, -I + 1, \ldots, I - 1, I$.

Let us assume that the polarization of the incident radiation is such that only one sublevel is excited, i.e., $l' = m'$. Then it follows from (27) that the intensity of radiation is modulated with a frequency $|l - m|\omega_2$. The angular dependence of the distribution of radiated light is included in the matrix elements of the emission operator C_{ml}, whereas the rotation matrices contain the dependence of the intensity on the magnitude of the magnetic fields. Of course they exhibit resonance behavior near $\delta_J = \delta_I = 0$. The term

$$[\Gamma + i(\mu' - \mu)\gamma_J H^J_{eff} + i(\zeta' - \zeta)\gamma_I H_{eff}]^{-1}$$

shows resonance near $\delta_J = 0$ and $\delta_I = 0$. If $\mu' = \mu$ and $\zeta' \neq \zeta$, this term shows resonance only for $\delta_I = 0$. Thus the light intensity reradiated by the atoms depends upon the resonance conditions for the nuclear system. When not one but several levels are excited, new phenomena may be observed, associated with the appearance of other modulation frequencies and with the change in the resonance conditions.

3. APPLICATION OF THE THEORY TO CADMIUM ATOMS

The results derived in the first two sections can be experimentally verified on, for example, such a system as gaseous cadmium. Owing to the non-zero nuclear moment ($I = 1/2$) of the cadmium atom, there are two sublevels in the 5^1S_0 state, characterized by the quantum numbers $m_0 = 0$ and $n_0 = \pm 1/2$, and six sublevels in the excited 5^3P_1 state, with quantum numbers $m = 0, \pm 1$ and $n = \pm 1/2$. The optical transitions (λ 3261 Å) are induced by light from a cadmium lamp. For light which is passed, say, through a linear polarizer, the matrix elements of the excitation

$$P_{mm_0} = e^0\langle m|D|m_0\rangle = P_m,$$

are equal to, according to [4],

$$P_{\pm 1} = \mp 2^{-1/2} D(\cos\alpha_j \cos\theta_j \mp \sin\alpha_j)\exp(\mp i\varphi_j),$$

$$P_0 = -D(\cos\alpha_j \sin\theta_j), \tag{28}$$

where $D = \langle 1|D|0\rangle$. The angles θ_j and φ_j define the direction of propagation of the light, and α_j is the angle between the electric vector and the unit vector associated with θ [4].

Using the explicit form of rotation matrices[5], the excitation matrices for light propagated along the z-axis and exciting a single sublevel in the excited state, we obtain

$$S = S_0[A\sin^2\theta + B(\cos^2\theta - 1/2\sin^2\theta) + \sin\theta\cos\theta \times \{C\cos(\omega_2 t - \varphi) + D\sin(\omega_2 t - \varphi)\} + F], \tag{29}$$

where the F are terms modulated with a frequency $2\omega_2$;

$$A = (1 + \cos^2\beta)/\Gamma,$$

$$B = \frac{\Omega_J^2(\Gamma^2 + 4\delta_J^2 + \Omega_J^2)}{(\Gamma^2 + \Omega_J^2 + \delta_J^2)(4\Omega_J^2 + 4\delta_J^2 + \Gamma^2)}A, \tag{30}$$

$$C = -\frac{\delta_J\Omega_J(4\delta_J^2 - 2\Omega_J^2 + \Gamma^2)}{(\Gamma^2 + \Omega_J^2 + \delta_J^2)(4\Omega_J^2 + 4\delta_J^2 + \Gamma^2)}A,$$

$$D = -\frac{\Gamma\Omega_J(4\delta_J^2 + \Omega_J^2 + \Gamma^2)}{(\Gamma^2 + \Omega_J^2 + \delta_J^2)(4\Omega_J^2 + 4\delta_J^2 + \Gamma^2)}A. \tag{31}$$

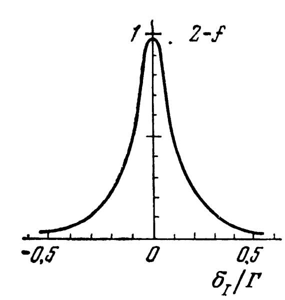
β is the angle between the effective magnetic field of the nuclear system and the constant magnetic field, as determined by the relation:

$$\sin\beta = \Omega_I(\Omega_I^2 + \delta_I^2)^{-1/2}. \tag{32}$$

In calculating the coefficients A, B, C, and D we omitted those terms which are very small in comparison with $c\cos^2\beta$.

The expression derived for the intensity of radiation propagated in a direction determined by the angles θ and φ not only depends on the resonance conditions in the electron system, but also changes appreciably when nuclear resonance is approached. The dependence of the intensity on the frequencies of the radio-frequency field is included in the function $1 + \cos^2\beta = f$, which enters as a factor in the coefficients A, B, C, and D. If we take $\Gamma = 0.4 \times 10^6$ cps for the cadmium atom[6], and $\gamma_I/2\pi = 903$ cps[7], then, with $\Omega_I/\Gamma = 0.01$, the dependence of the function $2 - f$ on the frequency difference δ_I/Γ will have the appearance shown in the figure. This dependence exhibits a strong resonance character when approaching the resonance condition $\delta_I = 0$. The half-width at half maximum is about $0.1\,\delta_I/\Gamma$, approximately 4×10^3 cps. Thus, the intensity curve duplicates the line shape of the nuclear resonance and may therefore serve to detect it. Besides this, clearly, new possibilities of determining nuclear g-factors and hyperfine splitting constants experimentally have been uncovered.

The dependence of the reradiated light intensity



Dependence of the fluorescent radiation intensity on the magnitude of the frequency difdifference δ_I/Γ.

on the magnitude of the microwave field is derived in the same way as in [3]. The dependence of the intensity on the size of the radio-frequency field is determined by (30). In the units shown this dependence has a Lorentzian form.

At the beginning of this section the assumption was made that the incident radiation causes resonance transitions among sublevels whose magnetic quantum numbers are equal to zero, i.e., one single electron sublevel of the 5^3P_1 state is excited. If not one but, say, three sublevels are excited, then by studying the spectrum of the total radiation, it is possible to derive a complete picture of the hyperfine splitting of the cadmium 5^3P_1 level. In conclusion one can note that this method for investigating nuclear and atomic phenomena suggests the well known ENDOR[8] technique of electron-nuclear resonance. However, unlike [8], in the case considered here the information on resonance phenomena in a nuclear system is contained in the atom's optical radiation.

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Translated by L. Marquet
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