Quadrupole Contribution in Semiclassical Radiation Theory*

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Within the frame-work of semiclassical theory two-level approximation in atomic system has been considered. Model proposed by M.D. Crisp and E.T. Jaynes has been modified. It is here shown that the time-dependent frequency shift depends on the higher multipole moments, retained in the Taylor expansion of electromagnetic field.

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1. INTRODUCTION

A general treatment of the interaction of photon with micro-particles is beyond the scope of quantum mechanics (QM). It is treated in quantum electrodynamics (QED), invoking additional principles concerning the laws of occurrence and disappearance of electromagnetic field. According to QM, an atom should remain in an excited state for long in absence of an external field, whereas experiments show that an atom transforms into normal state emitting a photon. It has been suggested that this limitation of QM could be explained taking into account the fact that the moving electron creates an electromagnetic field which reacts on the electron. Several authors considered this reverse action of field on electron several ways. One of these methods was proposed by Jaynes and Cummings [1] in 1963 where they clarified the relationship between the quantum theory of radiation, where electromagnetic field-expansion coefficients satisfy commutation relations, and semiclassical theory, where the electromagnetic field is considered as a definite function of time rather than as an operator.

An improved form of the semiclassical radiation theory was developed by Jaynes and Stroud [2] which includes the effect of the atom's radiation field back on the atom. Further Crisp and Jaynes [3] showed that, in the absence of an applied field, semiclassical theory predicts that an atom will decay spontaneously from an excited state with an characteristic time equal to the reciprocal of the Einstein A coefficient for the transition. The semiclassical radiation theory was also studied and developed by Berman [4], Salmon [5], Bosanac [6], Boudet [7] and many others.

In classical electrodynamics the radiative process are calculated from the self-energy of the electron in external fields. In contrast, in QED the self-energy is first thrown away and one begins with bare particles; then the self-energy is put back in *photon by photon*, hence the use of perturbation theory. Recently, Barut and coauthors developed a QED based on self-energy [8, 9] which gives the Lamb shift in semiclassical theory. Beside developing the theory by Barut and Van Huele [9] Blaive and Boudet [10, 11] have proposed a new method of calculation of the Lamb shift.

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Since the radiation of excited atoms is a fundamental process of radiation-matter interaction, which is responsible for important phenomena such as laser radiation, and spontaneous emission is an ubiquitous, particularly prominent and literally visible phenomenon, the problem in question has been studied by many authors from many other points of view. Kazaku and co authors have studied the spontaneous emission in circular cylindrical cavities [12, 13], while Cavalleri [14] considered a system where electrons are assumed to have the speed of light and electromagnetic self-reaction perpendicular to the velocity so that they perform a circular motion (spin or real *zitterbewegung*) that generates the zero-point field, hence special relativity. Cavalleri suggests that there exists zero-point fields in the surrounding space. The moving electron interacts with it and this explains the doubling of the gyromagnetic ratio. A QED treatment of radiative corrections in atoms was presented by Ackerhalt and Eberly [15]. For a detailed study of spontaneous emission from practical point of view one may consult [16] and references therein.

As we have already mentioned, we confine our study within the scope of semiclassical theory suggested by Jaynes and Cummings [1] and further developed by Crisp [3, 17]. The authors of the previously mentioned papers mainly confined their study within electric dipole moments. Here we make an attempt to enlarge this study taking into account the moments of higher order, particularly electric quadrupole moment.

2. FUNDAMENTAL EQUATIONS

Let us consider a nonrelativistic, spinless particle in external magnetic field. It can be described by the Hamiltonian

$$\hat{H} = \frac{1}{2m}\left[\hat{\mathbf{p}} - \frac{e}{c}\mathbf{A}\right]^2 - \frac{e^2}{r}. \tag{2.1}$$

Varying this Hamiltonian (2.1) with respect to $\mathbf{A}$ in account of the continuity equation

$$\frac{\partial \rho}{\partial t} + \mathrm{div}\mathbf{j} = 0, \tag{2.2}$$

one finds the current and charge density of the field

$$\mathbf{j} = \frac{ie\hbar}{2m}\{\Psi\nabla\Psi^* - \Psi^*\nabla\Psi\} - \frac{e^2}{mc}\mathbf{A}\Psi\Psi^*, \tag{2.3}$$

$$\rho = e\Psi^*\Psi. \tag{2.4}$$

Now, any state of atomic system may be expressed as

$$\Psi(\mathbf{r}, t) = \sum_\alpha a_\alpha(t)\psi_\alpha(\mathbf{r}) \tag{2.5}$$

where $\psi(\mathbf{r})$ is the eigenfunctions of $\hat{H}_0 = -(\hbar^2/2m)\nabla^2 - e^2/r$, i.e.,

$$\hat{H}_0\psi_\alpha(\mathbf{r}) = E_\alpha\psi_\alpha(\mathbf{r}). \tag{2.6}$$

Substituting (2.5) into (2.3) we obtain

$$\mathbf{j}(t,\mathbf{r}) = \frac{e\hbar}{2mi}\sum_{\alpha,\beta}\left[\rho_{\alpha\beta}\psi^*_\beta\nabla\psi_\alpha - \rho_{\beta\alpha}\psi_\beta\nabla\psi^*_\alpha\right]$$
$$-\frac{e^2}{mc}\mathbf{A}\sum_{\alpha,\beta} a_\alpha(t)a_\beta(t)\psi_\alpha(\mathbf{r})\psi_\beta(\mathbf{r}), \tag{2.7}$$

where

$$\rho_{\beta\alpha}(t) = a_\alpha(t)a^*_\beta(t) = \rho_{\alpha\beta}(t)^* \tag{2.8}$$

is the $\beta\alpha$ density matrix element of the atom in the Schrödinger picture that evolves according to

$$i\hbar\dot{\rho}_{\alpha\beta}(t) = \sum_\gamma[\hat{H}_{\alpha\gamma}\rho_{\gamma\beta} - \rho_{\alpha\gamma}\hat{H}_{\gamma\beta}]. \tag{2.9}$$

Taking the field to be weak we further neglect the diamagnetic term in the Hamiltonian and current density.

In the Coulomb gauge (div$\mathbf{A} = 0$) the equation for $\mathbf{A}$ is

$$\nabla^2\mathbf{A} - \frac{1}{c^2}\frac{\partial^2\mathbf{A}}{\partial t^2} = -\frac{4\pi}{c}\mathbf{j}^\perp, \tag{2.10}$$

where $\mathbf{j}^\perp$ is the transverse current density defined as

$$\mathbf{j}^\perp = \frac{1}{4\pi}\nabla\times\nabla\times\int\frac{\mathbf{j}(t,\mathbf{x}')}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}'. \tag{2.11}$$

Further we denote $\mathbf{j}^\perp = \mathbf{j}$. The solution to the Maxwell equation can be written as

$$\mathbf{A}(\mathbf{x},t) = \frac{1}{c}\int\frac{\mathbf{j}(\mathbf{x}',t-|\mathbf{x}-\mathbf{x}'|/c)}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}'. \tag{2.12}$$

Expanding $\mathbf{j}$ as a function of time in Eqn. (2.12) we obtain

$$\mathbf{A}(\mathbf{x},t) \approx \int\frac{j(\mathbf{x}',t)}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}' - \frac{1}{c}\int\frac{\partial j(\mathbf{x}',t)}{\partial t}d^3\mathbf{x}' + \frac{1}{2c^2}\int\frac{\partial^2 j(\mathbf{x}',t)}{\partial t^2}|\mathbf{x}-\mathbf{x}'|d^3\mathbf{x}' + \cdots . \tag{2.13}$$

Further expanding $|\mathbf{x}-\mathbf{x}'|$ for $x' \ll x$ one finds

$$\mathbf{A}(\mathbf{x},t) \approx \int\frac{j(\mathbf{x}',t)}{|\mathbf{x}-\mathbf{x}'|}d^3\mathbf{x}' - \frac{1}{c}\int\frac{\partial j(\mathbf{x}',t)}{\partial t}d^3\mathbf{x}' + \frac{x}{2c^2}\int\frac{\partial^2 j(\mathbf{x}',t)}{\partial t^2}d^3\mathbf{x}'$$
$$- \frac{x}{2c^3 r}\int\frac{\partial^2 j(\mathbf{x}',t)}{\partial t^2}x' d^3\mathbf{x}' - \cdots . \tag{2.14}$$

Substituting Eqn. (2.7) into Eqn. (2.14) and retaining the electric dipole and quadrupole moments we find

$$\mathbf{A}(\mathbf{x},t) \approx \sum_{\alpha\beta}\rho_{\alpha\beta}(t)\Big[\frac{-ie\hbar}{2\pi^2 mc}\int_0^\infty dk\int d\Omega(\beta|e^{-ik\cdot x'}\nabla|\alpha)_\perp e^{ik\cdot x}$$
$$+ \left(\frac{2}{3c^2}\Omega_{\alpha\beta} + \frac{ir}{3c^3}\Omega^3_{\alpha\beta}\right)D^{(1)}_{\alpha\beta} - \frac{ix_\alpha}{2c^3 r}\Omega^3_{\alpha\beta}D^{(2)}_{\alpha\beta}\Big] + \mathbf{A}_0(\mathbf{x},t), \tag{2.15}$$

where the transition frequencies and the electric dipole and quadrupole moments are defined, respectively, as

$$\Omega_{\alpha\beta} = (E_\alpha - E_\beta)/\hbar, \tag{2.16}$$

$$\mathbf{D}_{\alpha\beta} = \int \psi_\alpha e\mathbf{x}\psi^*_\beta d\mathbf{x}, \quad \text{or in components} \quad D^{(i)}_{\alpha\beta} = \int \psi_\alpha e x^i \psi^*_\beta d\mathbf{x}, \tag{2.17}$$

$$Q^{(ij)}_{\alpha\beta} = \int \psi_\alpha e r^{ij}\psi^*_\beta d\mathbf{x}, \qquad r^{ij} = \frac{1}{2}(x^i x^j - \frac{1}{3}r^2\delta^{ij}), \quad r = |\mathbf{x}|. \tag{2.18}$$

Here $\mathbf{A}_0$ is an externally applied field. Substituting Eqn. (2.15) into (2.9), for the density matrix we find

$$\begin{aligned}\dot\rho_{\alpha\beta} &= -i\Omega_{\alpha\beta}\rho_{\alpha\beta} - i\sum_\kappa(\Gamma_{\alpha\kappa} - \Gamma_{\kappa\beta})\rho_{\kappa\kappa}\rho_{\alpha\beta} \\ &- \sum_\kappa\bigl[\frac{1}{2}(\mathcal{A}_{\alpha\kappa} + \mathcal{A}_{\beta\kappa}) - (\mathcal{B}_{\alpha\kappa} + \mathcal{B}_{\beta\kappa}) + (\mathcal{C}_{\alpha\kappa} + \mathcal{C}_{\beta\kappa})\bigr]\rho_{\kappa\kappa}\rho_{\alpha\beta} \\ &- \frac{\mathbf{A}_0(0,t)}{\hbar c}\sum_\kappa\bigl[\Omega_{\alpha\kappa}\mathbf{D}_{\alpha\kappa}\rho_{\kappa\beta} - \Omega_{\kappa\beta}\mathbf{D}_{\kappa\beta}\rho_{\alpha\kappa}\bigr],\end{aligned} \tag{2.19}$$

where we define

$$\Gamma_{\alpha\beta} \equiv -\frac{e^2\hbar}{2\pi^2 m^2 c^2}\int_0^\infty\int d\Omega(\alpha|e^{ik\cdot x'}|\beta)_\perp(\beta|e^{-ik\cdot x}|\alpha)_\perp = \Gamma_{\beta\alpha}, \tag{2.20}$$

$$\mathcal{A}_{\alpha\beta} \equiv \frac{4}{3}(\mathbf{D}_{\alpha\beta}\mathbf{D}_{\beta\alpha}/\hbar c^3)\Omega^3_{\alpha\beta} = -\mathcal{A}_{\beta\alpha}, \quad \text{Einstein coefficient}, \tag{2.21}$$

$$\mathcal{B}_{\alpha\beta} \equiv (\mathbf{D}_{\alpha\beta}\Delta_{\alpha\beta}/\hbar c^4)\Omega^3_{\alpha\beta} \equiv, -\mathcal{B}_{\beta\alpha}, \qquad \Delta_{\alpha\beta} = \int r\bar J_{\alpha\beta}(\mathbf{x})d\mathbf{x}, \tag{2.22}$$

$$\mathcal{C}_{\alpha\beta} \equiv (Q^{ij}_{\alpha\beta}\delta^k_{\alpha\beta}/\hbar c^4)\Omega^3_{\alpha\beta} \equiv -\mathcal{C}_{\beta\alpha}, \qquad \delta^k_{\alpha\beta} = \int \frac{x^k}{r}\bar J_{\alpha\beta}(\mathbf{x})d\mathbf{x}. \tag{2.23}$$

Here we denote $\bar J_{\alpha\beta} = (e\hbar/2mi)\bigl[\psi^*_\beta\nabla\psi_\alpha - \psi_\beta\nabla\psi^*_\alpha\bigr]$.

The equation (2.19) can be written in the following way where the repeating index denotes summation

$$\begin{aligned}\dot\rho_{\alpha\beta} &= -i\Omega_{\alpha\beta}\rho_{\gamma\tau}M_{\alpha\beta\gamma\tau} - i(\Gamma_{\alpha\kappa} - \Gamma_{\kappa\beta})\rho_{\kappa\kappa}\rho_{\gamma\tau}M_{\alpha\beta\gamma\tau} \\ &- \bigl[\frac{1}{2}(\mathcal{A}_{\alpha\kappa} + \mathcal{A}_{\beta\kappa}) - (\mathcal{B}_{\alpha\kappa} + \mathcal{B}_{\beta\kappa}) + (\mathcal{C}_{\alpha\kappa} + \mathcal{C}_{\beta\kappa})\bigr]\rho_{\kappa\kappa}\rho_{\gamma\tau}M_{\alpha\beta\gamma\tau} \\ &- \frac{A_0(0,t)}{\hbar c}\bigl[\Omega_{\alpha\kappa}D^{(1)}_{\gamma\kappa}\rho_{\kappa\tau} - \Omega_{\kappa\beta}D^{(1)}_{\kappa\tau}\rho_{\gamma\kappa}\bigr]M_{\alpha\beta\gamma\tau},\end{aligned} \tag{2.24}$$

where we define $M_{\alpha\beta\gamma\tau} = \delta_{\alpha\gamma}\delta_{\beta\tau}$.

As one sees from (2.19) or (2.24), the off-diagonal density matrix elements oscillate at frequencies $\Omega_{\alpha\beta} + \delta\Omega_{\alpha\beta}(t)$, where the time-dependent frequency-shift is

$$\delta\Omega_{\alpha\beta}(t) = -\sum_\kappa(\Gamma_{\alpha\kappa} - \Gamma_{\kappa\beta})\rho_{\kappa\kappa}(t). \tag{2.25}$$

Now the expectation of dipole moment of the atom

$$< \mu >= \int \Psi^*(\mathbf{x}, t) e\mathbf{x}\Psi(\mathbf{x}, t) d\mathbf{x} \tag{2.26}$$

in account of (2.5) can be written as

$$< \mu >= \sum_{\alpha\beta} \mathbf{D}_{\alpha\beta}\rho_{\beta\alpha}(t). \tag{2.27}$$

Thus we see that the off-diagonal matrix elements are directly connected with the expectation of dipole moment.

3. SPONTANEOUS DECAY

In what follows we take into account only two of these levels. We choose the zero from which we measure the energies to be midway between the two active levels, so that

$$E_2 = -E_1. \tag{3.1}$$

The equation (2.24) can then be written as

$$\dot\rho_{11} = -2q\rho_{11}\rho_{22}, \tag{3.2}$$
$$\dot\rho_{22} = 2q\rho_{11}\rho_{22}, \tag{3.3}$$
$$\dot\rho_{12} = -i\big[\Omega_{12} + \Gamma_{11}\rho_{11} - \Gamma_{22}\rho_{22} - \Gamma_{12}(\rho_{11} - \rho_{22})\big]\rho_{12} + q(\rho_{11} - \rho_{22})\rho_{12}, \tag{3.4}$$
$$\dot\rho_{21} = -i\big[\Omega_{21} - \Gamma_{11}\rho_{11} + \Gamma_{22}\rho_{22} + \Gamma_{12}(\rho_{11} - \rho_{22})\big]\rho_{21} + q(\rho_{11} - \rho_{22})\rho_{21}, \tag{3.5}$$

where we denote $2q = \mathcal{A}_{12} - 2\mathcal{B}_{12} + 2\mathcal{C}_{12}$.

Let us now rewrite $\rho_{\alpha\beta}$ in the form [18, 19]

$$\rho_{\alpha\beta} = \frac{1}{2}\big(\delta_{\alpha\beta} + P_j\sigma^j_{\alpha\beta}\big), \tag{3.6}$$

where σ^j are the Pauli matrices and $\mathbf{P} = (P_x, P_y, P_z)$ is a unit vector of three-dimensional Poincaré representation. It follow from (3.6):

$$\begin{aligned} \rho_{11} &= \frac{1}{2}\big(1 + P_z\big), \quad \rho_{12} = \frac{1}{2}\big(P_x - iP_y\big), \\ \rho_{22} &= \frac{1}{2}\big(1 - P_z\big), \quad \rho_{21} = \frac{1}{2}\big(P_x + iP_y\big), \end{aligned} \tag{3.7}$$

or equivalently,

$$\rho_{11} + \rho_{22} = 1, \quad \rho_{11} - \rho_{22} = P_z, \quad \rho_{12} + \rho_{21} = P_x, \quad \rho_{12} - \rho_{21} = -iP_y. \tag{3.8}$$

In account of (3.7) and (3.8) from (3) we find the following system of equations

$$\dot P_x = qP_zP_x + (\Omega_{12} + \tau + \lambda P_z)P_y, \tag{3.9}$$
$$\dot P_y = qP_zP_y - (\Omega_{12} + \tau + \lambda P_z)P_x, \tag{3.10}$$
$$\dot P_z = q(P_z^2 - 1), \tag{3.11}$$

where we denote $\tau = (\Gamma_{11} - \Gamma_{22})/2$ and $\lambda = (\Gamma_{22} + \Gamma_{11})/2 - \Gamma_{12}$. The solutions to the system of equations (3) read

$$\begin{aligned} P_x &= \cos\left[\Omega_{12}(t-t_0) + \tau(t-t_0) + (\lambda/q)\ln\cosh q(t-t_0)\right] \mathrm{sech}\, q(t-t_0), && (3.12)\\ P_y &= \sin\left[\Omega_{12}(t-t_0) + \tau(t-t_0) + (\lambda/q)\ln\cosh q(t-t_0)\right] \mathrm{sech}\, q(t-t_0), && (3.13)\\ P_z &= -\tanh q(t-t_0), && (3.14) \end{aligned}$$

Rewriting (3) in terms of ρ we find

$$\begin{aligned} \rho_{11} &= 1/\Big[\exp\left[2q(t-t_0)\right] + 1\Big], && (3.15)\\ \rho_{22} &= 1/\Big[\exp\left[-2q(t-t_0)\right] + 1\Big], && (3.16)\\ \rho_{12} &= \Big[\exp\Big(-i[\Omega_{12}(t-t_0) + \tau(t-t_0) + (\lambda/q)\ln\cosh q(t-t_0)]\Big)\Big] \mathrm{sech}\, q(t-t_0), && (3.17)\\ \rho_{21} &= \Big[\exp\Big(i[\Omega_{12}(t-t_0) + \tau(t-t_0) + (\lambda/q)\ln\cosh q(t-t_0)]\Big)\Big] \mathrm{sech}\, q(t-t_0). && (3.18) \end{aligned}$$

The Eqn. (3.16) predicts a nonexponential decay for an atom in its excited state. This corresponds to a fundamental difference between semiclassical theory and QED.

For the expectation value of the energy in account of (3.1) we find

$$\begin{aligned} < H_0 > &= E_1\rho_{11}(t) + E_2\rho_{22}(t) = -\frac{\hbar}{2}\Omega_{21}(\rho_{22} - \rho_{11})\\ &= -\frac{\hbar}{2}\Omega_{21}\tanh[q(t-t_0)], && (3.19) \end{aligned}$$

where as, for the expectation of the dipole moment we obtain

$$\begin{aligned} < \mu > &= \mathbf{D}_{21}(\rho_{12} + \rho_{21}) = \mathbf{D}_{21}P_x\\ &= \mathbf{D}_{21}\,\mathrm{sech}\, q(t-t_0)\cos\left[\Omega_{21}t + \vartheta(t)\right], && (3.20) \end{aligned}$$

where we define

$$\vartheta(t) = \vartheta_0 - \tau t - (\lambda/q)\ln\cosh q(t-t_0), \quad \vartheta_0 = [(\Gamma_{11} - \Gamma_{22})/2 - \Omega_{21}]t_0, \qquad (3.21)$$

which corresponds to a time-dependent frequency shift

$$\delta\Omega_{21}(t) = d\vartheta/dt = -\tau - \lambda\tanh q(t-t_0) \qquad (3.22)$$

Comparing (3.22) with those obtained in [3] one finds the additional frequency shift as

$$\Delta(\delta\Omega_{21}(t)) = \lambda\frac{\tanh[(\mathcal{C}_{21} - \mathcal{B}_{21})(t-t_0)]\mathrm{sech}^2[\mathcal{A}_{21}(t-t_0)/2]}{1 + \tanh[\mathcal{A}_{21}(t-t_0)/2]\tanh[(\mathcal{C}_{21} - \mathcal{B}_{21})(t-t_0)]}. \qquad (3.23)$$

Thus we see that higher multipole moments, in particular quadrupole one, contribute to the spontaneous decay of the atom from an exited state beside Einstein A coefficient.

4. CONCLUSION

Using the semiclassical formulation of radiation theory, the frequency shift due to the quadrupole term in the external magnetic field has been calculated. It has been shown that the radiation damping due to quadrupole radiation is non-trivial [cf. Eqn. (3.23)] though is much smaller than the one due to dipole radiation.

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