Quadrupole Contribution in Semiclassical Radiation Theory

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Within the framework 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 has been 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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Issues, related to the problem of interaction of photon and micro-particle, in their full length are beyond the scope of quantum mechanics. They cannot be considered without invoking additional principles concerning the laws of occurrence and disappearance of electromagnetic field. According to quantum mechanics atom should remain in excited state for long in absence of external field, whereas experiment shows that atom transforms into normal state emitting photon. This contradiction can be explained if we take into account the fact that the moving electron creates electromagnetic field which acts on the electron. Several authors tried to consider this reverse action of field on electron several ways. One of these methods was proposed by Jaynes and Cummings [1] in 1963 that was further developed by Jaynes and Cot and many others [2–7].

In classical electrodynamics the radiative process are calculated from self-energy of the electron in external fields. In contrast, in quantum electrodynamics, 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 quantum electrodynamics based on self-energy [8,9].

Authors of the papers mentioned previously mainly confined their study within electric dipole moment. Here we make an attempt to enlarge this study taking into account the moments of higher order, particularly electric quadrupole moment.

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

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

Varying this Hamiltonian with respect to \( \vec{A} \) and using the continuity equation \( \frac{\partial \rho}{\partial t} + \text{div} \vec{j} = 0 \) one finds

\[
\vec{j} = \frac{i e \hbar}{2 m} \left\{ \Psi \nabla \Psi^* - \Psi^* \nabla \Psi \right\} - \frac{e^2}{mc} \vec{A} \Psi \Psi^* \tag{2}
\]

\[
\rho = e \Psi^* \Psi \tag{3}
\]

Taking the field to be weak one we further neglect the diamagnetic term in the Hamiltonian and current density. Now, any state of atomic system may be expressed as

\[
\Psi(\vec{r}, t) = \sum_{\alpha} a_{\alpha}(t) \psi_{\alpha}(\vec{r}) \tag{4}
\]

where \( \psi(\vec{r}) \) is the eigen functions of \( \hat{H}_0 = -\left(\hbar^2/2m\right) \nabla^2 - e^2/r \), i.e.,

\[
\hat{H}_0 \psi_{\alpha}(\vec{r}) = E_{\alpha} \psi_{\alpha}(\vec{r})
\]

Putting (4) into (3) we obtain

\[
\vec{j}(t, \vec{r}) = \frac{e \hbar}{2mi} \sum_{\alpha, \beta} \left[ \rho_{\alpha \beta} \psi_\beta \nabla \psi_\alpha - \rho_{\beta \alpha} \psi_\beta \nabla \psi_\alpha^* \right] \tag{5}
\]

where
Putting $\Psi = \sum_\gamma [\hat{\mathcal{H}}_{\alpha\gamma} \rho_{\gamma\beta} - \rho_{\alpha\gamma} \hat{\mathcal{H}}_{\gamma\beta}]$

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

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

Since the magnetic field obeys the Maxwell equations, for $\mathbf{A}$ in Coulomb gauge ($\text{div} \mathbf{A} = 0$) we can write

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

Here $\mathbf{j}$ is the transverse current density and defines as

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

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

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

Taylor expanding the expression for $\mathbf{j}$ one gets

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

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

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

Putting $\mathbf{A} = \sum_\alpha a_\alpha(t) \psi_\alpha(\mathbf{x}, t)$, where $\psi$: $\mathbf{H}_0 \psi_\alpha = E_\alpha \psi_\alpha$ into the equation above and retaining the electric dipole and quadrupole moments we find

$$\mathbf{A}(\mathbf{x}, t) \approx \sum_{\alpha\beta} \rho_{\alpha\beta}(t) \int \frac{e^{i\mathbf{k} \cdot \mathbf{x}'}}{2\pi^2 mc} \int_0^\infty d\Omega(\beta)e^{-ik \cdot \mathbf{x}'} \nabla|\alpha\rangle \langle \alpha| e^{ik \cdot \mathbf{x}}$$

$$+ \left( \frac{2}{3c^2} \Omega_{\alpha\beta} + \frac{i r}{3c^3} \Omega_{\alpha\beta}^3 \right) D^{(1)}_{\alpha\beta} - \frac{ix_{\alpha\beta}}{2c^3 r} \Omega_{\alpha\beta}^3 D^{(2)}_{\alpha\beta} + \mathbf{A}_0(\mathbf{x}, t)$$

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

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

$$\mathbf{D}_{\alpha\beta} = \int \psi_\alpha^* e^{i\mathbf{r} \cdot \mathbf{x}'} dx', \quad \text{or in components} \quad D^{(i)}_{\alpha\beta} = \int \psi_\alpha^* e^{i\mathbf{r} \cdot \mathbf{x}'} dx'$$

$$Q^{(ij)}_{\alpha\beta} = \int \psi_\alpha^* e^{i\mathbf{r} \cdot \mathbf{x}'} dx', \quad r^{ij} = \frac{1}{2}(x^i x^j - \frac{1}{3} \delta_{ij}), \quad r = |x|,$$

Here $\mathbf{A}_0$ is an externally applied field. Putting the expression for $\mathbf{A}$ into (15), for density matrix we find

$$\dot{\rho}_{\alpha\beta} = -i\Omega_{\alpha\beta} \rho_{\alpha\beta} - i \sum_\kappa (\Gamma_{\alpha\kappa} - \Gamma_{\kappa\beta}) \rho_{\kappa\kappa} \rho_{\alpha\beta}$$

$$- \sum_\kappa \left[ \frac{1}{2} (A_{\alpha\kappa} + A_{\beta\kappa}) - (B_{\alpha\kappa} + B_{\beta\kappa}) + (C_{\alpha\kappa} + C_{\beta\kappa}) \right] \rho_{\kappa\kappa} \rho_{\alpha\beta}$$

$$- \frac{\mathbf{A}_0(0, t)}{\hbar c} \sum_\kappa \left[ \Omega_{\alpha\kappa} \mathbf{D}_{\alpha\kappa} \rho_{\beta\beta} - \Omega_{\alpha\beta} \mathbf{D}_{\kappa\beta} \rho_{\alpha\kappa} \right],$$

where we define
Here we denote \( \bar{\sigma} \) where

\[
\Gamma_{\alpha\beta} = -\frac{e^2\hbar}{2m^2c^2} \int_0^\infty d\Omega(\alpha|e^{ikx'}|\beta)\bar{\Omega}(\alpha|e^{-ikx}|\beta) = \Gamma_{\beta\alpha},
\]  

(15a)

\[
A_{\alpha\beta} = \frac{4}{3}(D_{\alpha\beta}D_{\alpha\beta}/\hbar c^3)\Omega^3_{\alpha\beta} = -A_{\beta\alpha}, \quad \text{Einstein coefficient}
\]

(15b)

\[
B_{\alpha\beta} = (D_{\alpha\beta}A_{\alpha\beta}/\hbar c^4)\Omega^3_{\alpha\beta} = -B_{\beta\alpha}, \quad \Delta_{\alpha\beta} = \int r\bar{J}_{\alpha\beta}(x)dx,
\]

(15c)

\[
C_{\alpha\beta} = (Q_{\alpha\beta}^j\delta_{\alpha\beta}/\hbar c^4)\Omega^3_{\alpha\beta} = -C_{\beta\alpha}, \quad \delta_{\alpha\beta} = \int \frac{2k}{r}\bar{J}_{\alpha\beta}(x)dx.
\]

(15d)

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

The equation (14) can be written in the following way where the repeating index denotes summation

\[
\dot{\rho}_{\alpha\beta} = -i\Omega_{\alpha\beta}\rho_{\gamma\gamma}M_{\alpha\beta\gamma\gamma} - i(\Gamma_{\alpha\gamma} - \Gamma_{\beta\gamma})\rho_{\kappa\kappa}\rho_{\gamma\gamma}M_{\alpha\beta\gamma\gamma}
\]

\[
- [\frac{1}{2}(A_{\alpha\kappa} + A_{\beta\kappa}) - (B_{\alpha\kappa} + B_{\beta\kappa}) + (C_{\alpha\kappa} + C_{\beta\kappa})]\rho_{\kappa\kappa}\rho_{\gamma\gamma}M_{\alpha\beta\gamma\gamma}
\]

\[
- \frac{A_0(0, t)}{\hbar c}[\Omega_{\alpha\kappa}D_{\gamma\gamma}^{(1)}\rho_{\kappa\kappa} - \Omega_{\alpha\beta}D_{\kappa\kappa}^{(1)}\rho_{\beta\beta}]M_{\alpha\beta\gamma\gamma},
\]

(16)

where \( M_{\alpha\beta\gamma\tau} = \delta_{\alpha\gamma}\delta_{\beta\tau} \).

As one sees from (14) or (16), the off-diagonal density matrix elements oscillate at frequencies \( \Omega_{\alpha\beta} + \delta\Omega_{\alpha\beta}(t) \), where the time-dependent frequency-shift is

\[
\delta\Omega_{\alpha\beta}(t) = -\sum_\kappa (\Gamma_{\alpha\kappa} - \Gamma_{\beta\kappa})\rho_{\kappa\kappa}(t)
\]

(17)

Now the expectation of dipole moment of the atom

\[
< \mu > = \int \Psi^*(x, t)e^xe^\Psi(x, t)dx
\]

in account of (8) can be written as

\[
< \mu > = \sum_{\alpha\beta}D_{\alpha\beta}\rho_{\beta\alpha}(t).
\]

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

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

\[
E_2 = -E_1
\]

(18)

The equation (16) can then be written as

\[
\dot{\rho}_{11} = -2q\rho_{11}\rho_{22}
\]

\[
\dot{\rho}_{22} = 2q\rho_{11}\rho_{22}
\]

\[
\dot{\rho}_{12} = -i[\Omega_{12} + \Gamma_{11}\rho_{11} - \Gamma_{22}\rho_{22} - \Gamma_{12}(\rho_{11} - \rho_{22})]\rho_{12} + q(\rho_{11} - \rho_{22})\rho_{12}
\]

\[
\dot{\rho}_{21} = -i[\Omega_{21} - \Gamma_{11}\rho_{11} + \Gamma_{22}\rho_{22} + \Gamma_{12}(\rho_{11} - \rho_{22})]\rho_{21} + q(\rho_{11} - \rho_{22})\rho_{21}
\]

(19)

where we denote \( 2q = A_{12} - 2B_{12} + 2C_{12} \).

Let us now rewrite \( \rho_{\alpha\beta} \) in the form (17)

\[
\rho_{\alpha\beta} = \frac{1}{2}(\delta_{\alpha\beta} + P_j\sigma^j_{\alpha\beta})
\]

(20)

where \( \sigma^j \) are the Pauli matrices and \( P = (P_x, P_y, P_z) \) is a unit vector of three-dimensional Poincaré representation.

From (20) follow:
Thus we see that beside Einstein A to the spontaneous decay of the atom from an exited state.

\[
\rho_{11} = \frac{1}{2}(1 + P_z), \quad \rho_{12} = \frac{1}{2}(P_x - iP_y),
\]

\[
\rho_{22} = \frac{1}{2}(1 - P_z), \quad \rho_{21} = \frac{1}{2}(P_x + iP_y)
\]

or equivalently,

\[
\rho_{11} + \rho_{22} = 1, \quad \rho_{11} - \rho_{22} = P_z, \quad \rho_{12} + \rho_{21} = P_x, \quad \rho_{12} - \rho_{21} = -iP_y
\]

In account of (21) and (22) from (19) we find the following system of equations

\[
\dot{P}_x = qP_zP_x + (\Omega_{12} + \tau + \lambda P_z)P_y
\]

\[
\dot{P}_y = qP_zP_y - (\Omega_{12} + \tau + \lambda P_z)P_x
\]

\[
\dot{P}_z = q(P_z^2 - 1)
\]

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 (23) read

\[
P_x = \cos[\Omega_{12}(t - t_0) + \tau(t - t_0) + (\lambda/q)\ln \cosh q(t - t_0)] \text{sech} q(t - t_0)
\]

\[
P_y = \sin[\Omega_{12}(t - t_0) + \tau(t - t_0) + (\lambda/q)\ln \cosh q(t - t_0)] \text{sech} q(t - t_0)
\]

\[
P_z = -\tanh q(t - t_0)
\]

Rewriting (24) in terms of \(\rho\) we find

\[
\rho_{11} = 1/\left[\exp[2q(t - t_0)] + 1\right],
\]

\[
\rho_{22} = 1/\left[\exp[-2q(t - t_0)] + 1\right],
\]

\[
\rho_{12} = \left[\exp\left(-i(\Omega_{12}(t - t_0) + \tau(t - t_0) + (\lambda/q)\ln \cosh q(t - t_0))\right)\right] \text{sech} q(t - t_0),
\]

\[
\rho_{21} = \left[\exp\left(i(\Omega_{12}(t - t_0) + \tau(t - t_0) + (\lambda/q)\ln \cosh q(t - t_0))\right)\right] \text{sech} q(t - t_0).
\]

For the expectation value of the energy in account of (18) we find

\[
< H_0 > = E_1\rho_{11}(t) + E_2\rho_{22}(t) = -\frac{\hbar}{2}\Omega_{21}(\rho_{22} - \rho_{11})
\]

\[
= -\frac{\hbar}{2}\Omega_{21}\tanh[q(t - t_0)].
\]

\[
< \mu > = D_{21}(\rho_{12} + \rho_{21}) = D_{21}P_x
\]

\[
= D_{21}\text{sech} q(t - t_0) \cos[\Omega_{21}t + \phi(t)],
\]

where we define

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

and corresponds to a time-dependent frequency shift

\[
\delta\Omega_{21}(t) = d\phi/dt = -\tau - \lambda \tanh q(t - t_0)
\]

Comparing (24) with those obtained in (2) one finds the additional frequency shift as

\[
\Delta(\delta\Omega_{21}(t)) = \lambda \frac{\tanh[(C_{21} - B_{21})(t - t_0)]\text{sech}^2[A_{21}(t - t_0)/2]}{1 + \tanh[A_{21}(t - t_0)/2]\tanh[(C_{21} - B_{21})(t - t_0)]}
\]

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

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