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Modification of the Resonance Fluorescence Spectrum of a Two-Level Atom in the Near Field of a Plasmonic Nanoparticle

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The resonance fluorescence spectrum of an atom located in the near field of a plasmonic nanoparticle is considered. It is shown that, as the atom gets close to the nanoparticle, the high-frequency peak of the Mollow triplet disappears and the spectrum has a Fano resonance shape. The low-frequency peak also disappears as the distance between the atom and the nanoparticle decreases further. For small distances, when the atom interacts with the nanoparticle much more strongly than with the external field, the spectrum represents a Lorentzian line whose width is proportional to the square of the atom–nanoparticle coupling constant.

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1. INTRODUCTION

In connection with the rapid progress of plasmonics in recent years [1–5], much interest has been attracted to the problem of the influence of interaction between two-level atoms and plasmonic structures on their optical properties. For example, one can mention the dipole nanolaser [6, 7], spaser [8, 9], nanoscale light emitters [10, 11], and active metamaterials [12–19]. The starting point for these studies was the paper by Purcell et al. [20], after which the investigation of the effect of the environment on the spontaneous emission of an atom commenced (see [21] and references therein). In particular, the influence of nanostructures on the resonance fluorescence spectrum of atoms in the context of Purcell's approach was considered in [22, 23].

The resonance fluorescence spectrum (Mollow triplet) was measured for the first time in [24, 25] and was calculated theoretically in [26, 27]. This triplet is described by two parameters: the linewidth and atom–field coupling constant (Rabi frequency). The effect of a plasmonic nanoparticle on the resonance fluorescence spectrum of a two-level atom was first analyzed in [23], where both the modification of the atom radiation field caused by the presence of the nanoparticle and the change in the local field in the vicinity of the nanoparticle caused by the particle's reemission (i.e., the renormalization of the Rabi frequency) were taken into account.

In this approach, however, the phases of both atom and nanoparticle dipole moment oscillations are disregarded. The resulting interference may lead to a non-Lorentzian shape of the spectrum. The fluorescence spectrum can have a so-called Fano resonance shape (on the Fano resonance, see [28]). It should also be noted that, in the case of a small nanoparticle (≤20 nm), only a minor fraction of the external-field energy is reemitted, the major fraction being spent on Joule losses in the metal [29] (the characteristic nanoparticle nonradiative decay time is $\tau_a \sim 10^{-14}$ s [30]). For this reason, the number of plasmons (energy quanta of the nanoparticle near field [31]) excited in the nanoparticle becomes small or comparable to unity. In this case, quantum fluctuations of the near field of the nanoparticle should be taken into account.

Here, we consistently take into account the effect of quantum fluctuations and correlations on the dynamics of the nanoparticle and the two-level atom in the approximation of small plasmon number. We demonstrate that, as the atom gets close to the nanoparticle, the shape of the Mollow triplet becomes asymmetric owing to the Fano resonance. When the atom and nanoparticle are still nearer to each other, the side maxima disappear, so that the triplet degenerates into a single Lorentzian.

2. DYNAMICS OF AN ATOM IN THE FIELD OF AN EXTERNAL ELECTROMAGNETIC WAVE: RESONANCE FLUORESCENCE SPECTRUM

Let us consider the interaction of a single two-level atom with an external monochromatic electromagnetic wave $\mathbf{E}(\mathbf{r}, t) = \mathbf{E}_0 \cos \nu t$. The Hamiltonian of the system can be written as follows [32–34]:

$$\hat{H} = \hat{H}_{\mathrm{TLS}} + \hat{H}_{\mathrm{TLS}-w}, \tag{1}$$

where $\hat{H}_{\mathrm{TLS}} = \hbar\omega_{\mathrm{TLS}}\hat{\tilde{\sigma}}^{\dagger}\hat{\tilde{\sigma}}$ is the Hamiltonian of the two-level atom and the operator $\hat{V}_{\mathrm{TLS}-w} = -\hat{\mathbf{d}}_{\mathrm{TLS}}\mathbf{E}$ describes the interaction between the two-level atom and the field of the external wave. Here, $\hat{\mathbf{d}}_{\mathrm{TLS}} = \mu_{\mathrm{TLS}}\mathbf{e}_{\mathrm{tls}}[\hat{\tilde{\sigma}}(t) + \hat{\tilde{\sigma}}^{\dagger}(t)]$ is the dipole moment operator of the atom, where $\hat{\tilde{\sigma}} = |g\rangle\langle e|$ is the operator of the transition between the excited and the ground states of the atom ($|e\rangle$ and $|g\rangle$, respectively), $\mu_{\mathrm{TLS}} = \langle e|er|g\rangle$ is the magnitude of the dipole transition matrix element, and $\mathbf{e}_{\mathrm{TLS}}$ is the unit vector in the direction of the dipole moment. For simplicity, let us assume that the frequency of the external field coincides with the atomic transition frequency ($\omega_{\mathrm{TLS}} = \nu$). Substituting $\hat{\tilde{\sigma}} = \hat{\sigma}\exp(i\omega_a t)$ and neglecting antiresonance terms (the rotating-wave approximation [35]), one obtains the following expression for the interaction Hamiltonian: $\hat{V} = \hbar\Omega(\hat{\tilde{\sigma}}^{\dagger} + \hat{\tilde{\sigma}})/2$, where $\Omega = -\mathbf{E}\boldsymbol{\mu}_{\mathrm{TLS}}/\hbar$ is the Rabi frequency.

To describe the process of the atom relaxation, one can introduce a reservoir representing a set of bosonic modes (e.g., phonons) that interact with the atom and discard the interaction with this reservoir at a later stage [36]. As a result, the atom dynamics can be described by the following optical Bloch equations [34]:

$$\dot{\hat{D}} = i\Omega(\hat{\sigma} - \hat{\sigma}^{+}) - 2(\hat{D} + 1)/\tau_{\sigma} + \hat{F}_D(t), \tag{2}$$

$$\dot{\hat{\sigma}} = -\hat{\sigma}/\tau_{\sigma} + i\Omega\hat{D}/2 + \hat{F}_{\sigma}(t), \tag{3}$$

$$\dot{\hat{\sigma}}^{+} = -\hat{\sigma}^{+}/\tau_{\sigma} - i\Omega\hat{D}/2 + \hat{F}_{\sigma}^{+}(t), \tag{4}$$

here, the operator $\hat{D} = |e\rangle\langle e| - |g\rangle\langle g|$ represents the atomic level population inversion, τ_{σ} describes the atom relaxation, and operators $\hat{F}_D(T)$ and $\hat{F}_{\sigma}(t)$ represent the Markovian noise sources. The latter are characterized by zero average values $\langle\hat{F}_D(t)\rangle = \langle\hat{F}_{\sigma}(t)\rangle = 0$ and are δ-correlated, i.e., $\langle\hat{F}_{\sigma}(t)\hat{F}_{\sigma}^{+}(t')\rangle = (2/\tau_{\sigma})\delta(t - t')$ and $\langle\hat{F}_D(t)\hat{F}_D(t')\rangle = [4(1 + \langle\hat{D}\rangle)/\tau_{\sigma}]\delta(t - t')$ [34].

The spectrum of the atom can be determined from Eqs. (2)–(4) using the quantum regression theorem [26]. Here, we state this theorem in the form most convenient for the subsequent discussion [32, 37]. Let $\hat{X}_1, \hat{X}_2, \ldots, \hat{X}_n$ be a finite set of operators of a system that interacts with the reservoir so that noise in this system is Markovian and δ-correlated. The dynamics of the system is described by the following linear equations:

$$\dot{\hat{X}}_i = M_{ij}(t)\hat{X}_j + \xi_i + \hat{F}_i(t), \tag{5}$$

where the matrix M_{ij} and the free term ξ_i are determined by the properties of the system and the noise operator $\hat{F}_i(t)$ is δ-correlated, i.e., $\langle\hat{F}_i(t)\hat{F}_j(t')\rangle = 2D_{ij}\delta(t - t')$.

Then the theorem states that the average values of two-time correlation operators $\langle\hat{X}_i(t+\tau)\hat{X}_j(t)\rangle$ satisfy the following set of equations:

$$\begin{aligned}&\frac{\partial\langle\hat{X}_i(t+\tau)\hat{X}_j(t)\rangle}{\partial\tau}\\&= M_{ik}(\tau)\langle\hat{X}_k(t+\tau)\hat{X}_j(t)\rangle + \xi_i\langle\hat{X}_j(t)\rangle.\end{aligned} \tag{6}$$

Since Eqs. (2)–(4) represent a set of linear equations, we can immediately use the quantum regression theorem and obtain the following expression for the emission spectrum [33]:

$$\begin{aligned}S(\mathbf{r}, \omega) = {}&\frac{I_0(\mathbf{r})}{4\pi}\left[\frac{\Omega^2}{(2/\tau_{\sigma})^2 + 2\Omega^2}\right]\\&\times[s_1(\omega) + s_2(\omega) + s_3(\omega) + s_4(\omega)].\end{aligned} \tag{7}$$

Here, $I_0(\mathbf{r}) = \left(\dfrac{\omega_{\mathrm{TLS}}^2|\boldsymbol{\mu}_{\mathrm{TLS}}|\sin\eta}{c^2|\mathbf{r}|}\right)^2$ determines the dependence of the spectrum on the position of the observation point (where η is the angle between the z axis and the dipole, located in the xz plane, and r is the radius vector from the dipole to the observation point), and the frequency dependence is determined by the following four terms:

$$S_1(\omega) = \frac{4\pi(2/\tau_{\sigma})^2}{(2/\tau_{\sigma})^2 + 2\Omega^2}\delta(\omega_{\mathrm{TLS}} - \omega), \tag{8}$$

$$S_2(\omega) = \frac{(2/\tau_{\sigma})^2}{(\omega_{\mathrm{TLS}} - \omega)^2 + (1/\tau_{\sigma})^2}, \tag{9}$$

$$S_3(\omega) = \frac{A_{+}}{(\omega_{\mathrm{TLS}} + \mu - \omega)^2 + (3/2\tau_{\sigma})^2}, \tag{10}$$

$$S_4(\omega) = \frac{A_{-}}{(\omega_{\mathrm{TLS}} - \mu - \omega)^2 + (3/2\tau_{\sigma})^2}, \tag{11}$$

here, $\mu = (\Omega^2 - 1/4\tau_{\sigma}^2)^{1/2}$, $A_{\pm} = 3P/2\tau_{\sigma} \pm Q(\omega_{\mathrm{TLS}} \pm \mu - \omega)$, $P = [2\Omega^2 - (2\tau_{\sigma})^2]/[2\Omega^2 + (2/\tau_{\sigma})^2]$, and $Q = [10\Omega^2 - (1/\tau_{\sigma})^2]/2\mu\tau_{\sigma}[10\Omega^2 + (2/\tau_{\sigma})^2]$ [33]. One can see that the shape of the spectrum depends on the external field intensity. At low intensities, the dominant contribution comes from the Rayleigh scattering (term $s_1(\omega)$), which, for a strictly monochromatic external field, exhibits a δ-function behavior (the coherent contribution). For high intensities, the spectrum represents a Lorentzian line with a central peak (term $s_2(\omega)$) at the external-field frequency and two Lorentzian lines (terms $s_3(\omega)$ and $s_4(\omega)$) shifted from the central line by the Rabi frequency in the high-field limit; the widths of these three lines (which form the Mollow triplet) are determined by the atom relaxation rate.

Below we consider how the shape of this spectrum changes in the presence of a metallic nanoparticle in the vicinity of the atom. Thus, terms responsible for the dynamics of the near field of the nanoparticle will be taken into account in Hamiltonian (1).

3. HEISENBERG–LANGEVIN EQUATIONS FOR THE INTERACTING NANOPARTICLE AND TWO-LEVEL SYSTEM IN AN EXTERNAL FIELD

Now, let us consider the effect of a nanostructure, e.g., a metallic nanoparticle, on the resonance fluorescence spectrum of the atom. We assume that (i) the distance r between the atom and the nanoparticle satisfies the condition $kr \ll 1$, and, thus, their interaction can be described as the near-field interaction of two dipoles, and (ii) the atom transition frequency coincides with the plasmon-resonance frequency of the nanoparticle dipole mode. For describing the quantum dynamics of the nanoparticle and two-level atom in an external field, we use the following model Hamiltonian [12, 13, 31, 38]:

$$\hat{H} = \hat{H}_{\mathrm{NP}} + \hat{H}_{\mathrm{TLS}} + \hat{V}_{\mathrm{TLS\text{-}NP}} + \hat{V}_{\mathrm{TLS}-w} + \hat{V}_{\mathrm{NP}-w}. \quad (12)$$

Here, $\hat{H}_{\mathrm{HP}} = \hbar\omega_{\mathrm{SP}}\hat{\tilde{a}}^{+}\hat{\tilde{a}}$ is the Hamiltonian of the nanoparticle at the plasmon-resonance frequency ω_{SP}; $\hat{\tilde{a}}^{+}$ and $\hat{\tilde{a}}$ are the Bose creation and annihilation operators of the dipole surface plasmon, respectively [5, 8, 35]; and operators $\hat{V}_{\mathrm{NP}-w} = -\hat{\mathbf{d}}_{\mathrm{NP}}\hat{\mathbf{E}}$ and $\hat{V}_{\mathrm{TLS\text{-}NP}} = -\mathbf{d}_{\mathrm{TLS}}\mathbf{E}_{\mathrm{NP}}$ describe the interaction of the nanoparticle with the external monochromatic wave and the two-level atom, respectively.

The nanoparticle polarization at the plasmon-resonance frequency is described by the equation of an oscillator with a frequency equal to the plasmon-resonance frequency:

$$\ddot{\mathbf{d}}_{\mathrm{NP}} + \omega_{\mathrm{SP}}^2\mathbf{d}_{\mathrm{NP}} = 0. \quad (13)$$

Let us introduce Bose creation, $\hat{\tilde{a}}^{+}(t)$, and annihilation, $\hat{\tilde{a}}(t)$, operators of the dipole surface plasmon, satisfying the commutation relation $[\hat{\tilde{a}}(t), \hat{\tilde{a}}^{+}(t)] = 1$. Then, the dipole-moment operator can be written as

$$\hat{\mathbf{d}}_{\mathrm{NP}} = \tilde{\boldsymbol{\mu}}_{\mathrm{NP}}(\hat{\tilde{a}} + \hat{\tilde{a}}^{+})/\sqrt{2}, \quad (14)$$

and the harmonic-oscillator Hamiltonian is expressed as $\hat{H}_{\mathrm{SP}} = \hbar\omega_{\mathrm{SP}}\hat{\tilde{a}}^{+}\hat{\tilde{a}}$. In order to find $\tilde{\boldsymbol{\mu}}_{\mathrm{NP}}$, we compare the energy of a quantum and the energy of a single plasmon

$$\hbar\omega_{\mathrm{SP}} = \frac{1}{8\pi}\int_V \frac{\partial[\omega\,\mathrm{Re}\,\varepsilon(\omega)]}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}} \mathbf{E}_{\mathrm{NP}}\mathbf{E}_{\mathrm{NP}}^{*}dV. \quad (15)$$

In the case of a spherical nanoparticle, the electric field inside and outside the particle equals $\mathbf{E}_{\mathrm{in}} = -\tilde{\boldsymbol{\mu}}_{\mathrm{NP}}/r_{\mathrm{NP}}^3$ and $\mathbf{E}_{\mathrm{out}} = -\boldsymbol{\mu}_{\mathrm{NP}}/r^3 + 3(\tilde{\boldsymbol{\mu}}_{\mathrm{NP}} \cdot \mathbf{r})\mathbf{r}/r^5$, respectively. Let us take into account that

$$\begin{aligned}
&\frac{1}{8\pi}\int_V \frac{\partial[\omega\,\mathrm{Re}\,\varepsilon(\omega)]}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}} \mathbf{E}_{\mathrm{NP}} \cdot \mathbf{E}_{\mathrm{NP}}^{*}dV \\
&= \frac{1}{8\pi}\left\{\int \frac{\partial[\mathrm{Re}\,\varepsilon(\omega)]}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}} \mathbf{E}_{\mathrm{NP}} \cdot \mathbf{E}_{\mathrm{NP}}^{*}dV \right. \quad (16) \\
&\left. + \int_V \mathrm{Re}\,\varepsilon(\omega)|_{\omega_{\mathrm{SP}}}\mathbf{E}_{\mathrm{NP}} \cdot \mathbf{E}_{\mathrm{NP}}^{*}dV\right\}.
\end{aligned}$$

As a consequence of the Laplace equation $\nabla[\mathrm{Re}\,\varepsilon(\omega_{\mathrm{SP}})\nabla\varphi] = 0$ and the boundary conditions $\varepsilon_{\mathrm{in}}\frac{\partial\varphi_{\mathrm{in}}}{\partial\mathbf{r}} = \varepsilon_{\mathrm{out}}\frac{\partial\varphi_{\mathrm{out}}}{\partial\mathbf{r}}$, the second term in the braces vanishes:

$$\begin{aligned}
&\int_V \mathrm{Re}\,\varepsilon_{\mathrm{NP}}(\omega)|_{\omega_{\mathrm{SP}}}\mathbf{E}_{\mathrm{NP}} \cdot \mathbf{E}_{\mathrm{NP}}^{*}dV \\
&= \int_V \mathrm{Re}\,\varepsilon_{\mathrm{NP}}(\omega)|_{\omega_{\mathrm{SP}}}|\nabla\varphi|^2 dV \\
&= \left(\int_{V_{\mathrm{in}}} dV_{\mathrm{in}} + \int_{V_{\mathrm{out}}} dV_{\mathrm{out}}\right)\varphi^{*}\nabla[\mathrm{Re}\,\varepsilon_{\mathrm{NP}}(\omega)\nabla\varphi]|_{\omega_{\mathrm{SP}}} \quad (17) \\
&+ \int_{\partial V}\varphi^{*}\left(\varepsilon_{\mathrm{in}}\frac{\partial\varphi_{\mathrm{in}}}{\partial\mathbf{r}} - \varepsilon_{\mathrm{out}}\frac{\partial\varphi_{\mathrm{out}}}{\partial\mathbf{r}}\right)\mathbf{n} = 0.
\end{aligned}$$

In addition, we assume that the medium outside the nanoparticle has no dispersion, i.e., $\partial\varepsilon_{\mathrm{out}}/\partial\omega = 0$.

Then, we obtain the following expression for the plasmon energy:

$$W_{\mathrm{NP}} = \frac{1}{8\pi}\int_{V_{\mathrm{in}}} \omega \frac{\partial \mathrm{Re}\varepsilon_{\mathrm{NP}}(\omega)}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}} \mathbf{E}_{\mathrm{NP}}\cdot\mathbf{E}^{*}_{\mathrm{NP}} dV = \frac{|\tilde{\boldsymbol{\mu}}_{\mathrm{NP}}|^2}{6r^3_{\mathrm{NP}}}\omega\frac{\partial \mathrm{Re}\varepsilon_{\mathrm{NP}}(\omega)}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}}. \tag{18}$$

As a result,

$$\hbar\omega_{\mathrm{SP}} = \frac{|\tilde{\boldsymbol{\mu}}_{\mathrm{NP}}|^2}{6r^3_{\mathrm{NP}}}\omega_{\mathrm{SP}}\frac{\partial \mathrm{Re}\varepsilon_{\mathrm{NP}}(\omega)}{\partial\omega}\bigg|_{\omega_{\mathrm{SP}}}, \tag{19}$$

from whence $\tilde{\boldsymbol{\mu}}_{\mathrm{NP}} = \sqrt{6\hbar r^3_{\mathrm{NP}}/(\partial \mathrm{Re}\varepsilon_{\mathrm{NP}}/\partial\omega)}\mathbf{e}_{\mathrm{NP}}$. For convenience, let us designate $\boldsymbol{\mu}_{\mathrm{NP}} = \tilde{\boldsymbol{\mu}}_{\mathrm{NP}}/\sqrt{2}$. Thus, the particle dipole moment operator is $\hat{\mathbf{d}}_{\mathrm{NP}} = \boldsymbol{\mu}_{\mathrm{NP}}(\hat{\tilde{a}} + \hat{\tilde{a}}^{\dagger})$, where $\boldsymbol{\mu}_{\mathrm{NP}} = \sqrt{3\hbar r^3_{\mathrm{NP}}/(\partial \mathrm{Re}\varepsilon_{\mathrm{NP}}/\partial\omega)}\,\mathbf{e}_{\mathrm{NP}}$ (on the quantization procedure, see also [31]).

Thus, $\hat{V}_{\mathrm{TLS\text{-}NP}} = \hbar\Omega_{\mathrm{R}}(\hat{\tilde{a}}^{\dagger} + \hat{\tilde{a}})(\hat{\tilde{\sigma}}^{\dagger} + \hat{\tilde{\sigma}})$, where $\Omega_{\mathrm{R}} = [\boldsymbol{\mu}_{\mathrm{NP}}\boldsymbol{\mu}_{\mathrm{TLS}} - 3(\boldsymbol{\mu}_{\mathrm{TLS}}\cdot\mathbf{e}_r)(\boldsymbol{\mu}_{\mathrm{NP}}\cdot\mathbf{e}_r)]/\hbar r^3$ (here, $\mathbf{e}_r = \mathbf{r}/r$ is the unit vector) is the Rabi frequency for the interaction between the two-level atom and the nanoparticle.

Let us seek solutions of the form $\hat{\tilde{a}}(t) \equiv \hat{a}(t)\exp(-i\omega t)$ and $\hat{\tilde{\sigma}}(t) \equiv \hat{\sigma}(t)\exp(-i\omega t)$, where $\hat{a}(t)$ and $\hat{\sigma}(t)$ are the slowly varying amplitudes. Disregarding rapidly oscillating terms ~exp(±2$i\omega t$) (the rotating-wave approximation [35]), one can express the interaction Hamiltonian $\hat{V}_{\mathrm{TLS\text{-}NP}}$ in the Jaynes–Cummings form [33]:

$$\hat{V}_{\mathrm{TLS\text{-}NP}} = \hbar\Omega_{\mathrm{R}}(\hat{a}^{\dagger}\hat{\sigma} + \hat{\sigma}^{\dagger}\hat{a}). \tag{20}$$

Let the dipole moments of the two-level atom and the nanoparticle be collinear and oriented along the x axis. Then, the interaction between the nanoparticle and the external wave assumes the form

$$\hat{V}_{\mathrm{NP}-w} = \alpha\hbar\Omega(\hat{a}^{+} + \hat{a}), \tag{21}$$

where $\alpha = |\boldsymbol{\mu}|_{\mathrm{NP}}/|\boldsymbol{\mu}|_{\mathrm{TLS}}$.

The decay of the nanoparticle dipole moment can be described similarly to the decay of the dipole moment of an atom [39]. Starting from Hamiltonian (4), we obtain the following equations of motion describing the dynamics of the average values of the operators:

$$\langle\hat{D}\rangle = 2i\Omega_{\mathrm{R}}(\langle\hat{a}^{+}\hat{\sigma}\rangle - \langle\hat{\sigma}^{+}\hat{a}\rangle) + 2i\Omega(\langle\hat{\sigma}\rangle - \langle\hat{\sigma}^{+}\rangle) - 2(\langle\hat{D}\rangle + 1)/\tau_{\sigma}, \tag{22}$$

$$\langle\hat{\sigma}\rangle = -\langle\hat{\sigma}\rangle/\tau_{\sigma} + i\Omega_{\mathrm{R}}\langle\hat{a}\hat{D}\rangle + i\Omega\langle\hat{D}\rangle, \tag{23}$$

$$\langle\hat{\sigma}^{+}\rangle = -\langle\hat{\sigma}^{+}\rangle/\tau_{\sigma} - i\Omega_{\mathrm{R}}\langle\hat{a}^{+}\hat{D}\rangle - i\Omega\langle\hat{D}\rangle, \tag{24}$$

$$\langle\hat{a}\rangle = -\langle\hat{a}\rangle/\tau_{a} - i\Omega_{\mathrm{R}}\langle\hat{\sigma}\rangle - i\alpha\Omega, \tag{25}$$

$$\langle\hat{a}^{+}\rangle = -\langle\hat{a}^{+}\rangle/\tau_{a} + i\Omega_{\mathrm{R}}\langle\hat{\sigma}^{+}\rangle - i\alpha\Omega. \tag{26}$$

Equations (22)–(26) contain average values of the products of two operators. Using Hamiltonian (7), we can now write equations for these operator products. However, these equations will contain averages of the products of three and four operators, etc. Thus, there emerges an infinite chain of equations for the average values.

As was noted in the Introduction, owing to high Joule losses in metallic nanoparticles ($\tau_a \sim 10^{-14}$ s, so that $\Omega \ll \tau_a^{-1}$), the number of plasmons excited by the external field is smaller than unity: $\langle\hat{a}^{+}\hat{a}\rangle \sim (\alpha\Omega\tau_a)^2 \le 1$. Consequently, the average values of operator products containing plasmon creation or annihilation operators in the second or higher powers ($\langle\hat{\sigma}^{+}\hat{D}\hat{a}^{2}\rangle$, $\langle\hat{a}^{+}\hat{D}\hat{a}^{2}\rangle$, etc.) are also smaller than unity. Disregarding terms of the second and higher orders in the plasmon creation and annihilation operators, we obtain the following closed set of equations for the operator averages:

$$\langle\hat{\mathbf{x}}\rangle = M(\mathbf{x}) + \mathbf{x}_0. \tag{27}$$

Here,

$$\hat{\mathbf{x}} = (\hat{a}, \hat{a}^{+}, \hat{\sigma}, \hat{\sigma}^{+}, \hat{a}\hat{D}, \hat{a}^{+}\hat{D}, \hat{a}^{+}\hat{\sigma}\hat{a}, \hat{a}^{+}\hat{\sigma}^{+}\hat{a}, \hat{a}^{+}\hat{a}, \hat{D}, \hat{a}^{+}\hat{\sigma}, \hat{\sigma}^{+}\hat{a}, \hat{a}^{+}\hat{D}\hat{a})^{\mathrm{T}}, \tag{28}$$

$$\mathbf{x}_0 = (-i\alpha\Omega, i\alpha\Omega, 0, 0, 0, 0, 0, 0, 0, -2/\tau_{\sigma}, i\Omega_{\mathrm{R}}/2, -i\Omega_{\mathrm{R}}/2, 0)^{\mathrm{T}} \tag{29}$$

and

$$M = \begin{pmatrix} M_{11} & M_{12} & M_{13} \\ M_{21} & M_{22} & M_{23} \\ M_{31} & M_{32} & M_{33} \end{pmatrix}, \tag{30}$$

where

$$M_{11} = \begin{pmatrix} -1/\tau_a & 0 & -i\Omega_{\mathrm{R}} & 0 & 0 \\ 0 & -1/\tau_a & 0 & i\Omega_{\mathrm{R}} & 0 \\ 0 & 0 & -1/\tau_{\sigma} & 0 & i\Omega_{\mathrm{R}} \\ 0 & 0 & 0 & -1/\tau_{\sigma} & 0 \\ -2/\tau_{\sigma} & 0 & i\Omega_{\mathrm{R}} & 0 & -1/(\tau_{\sigma}/2 + \tau_a) \end{pmatrix}, \tag{31}$$

$$M_{22} = \begin{pmatrix} -2/(\tau_\sigma + 2\tau_a) & 0 & -2i\Omega & 0 & i\alpha\Omega \\ 0 & -2/(\tau_a + 2\tau_\sigma) & 0 & 0 & 0 \\ -i\Omega_R/2 & 0 & -2/(\tau_a + 2\tau_\sigma) & 0 & 0 \\ 0 & 0 & 0 & -2/\tau_a & 0 \\ 0 & 0 & 0 & 0 & -2/\tau_\sigma \end{pmatrix}, \tag{32}$$

$$M_{12} = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & i\Omega \\ -i\Omega_R & 0 & 0 & 0 & -i\Omega \\ 0 & 2i\Omega & 0 & 0 & -i\alpha\Omega \end{pmatrix}, \tag{33}$$

$$M_{21} = \begin{pmatrix} 0 & -2/\tau_\sigma & 0 & -i\Omega_R & 0 \\ i\Omega_R/2 & 0 & 0 & 0 & i\Omega_R/2 \\ 0 & -i\Omega_R/2 & 0 & 0 & 0 \\ i\alpha\Omega & -i\alpha\Omega & 0 & 0 & 0 \\ 0 & 0 & 2i\Omega & -2i\Omega & 0 \end{pmatrix}, \tag{34}$$

$$M_{13} = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & -i\Omega & 0 \end{pmatrix},$$

$$M_{23} = \begin{pmatrix} i\Omega & 0 & 0 \\ -i\alpha\Omega & 0 & i\Omega \\ 0 & i\alpha\Omega & -\Omega \\ -i\Omega_R & i\Omega_R & 0 \\ 2i\Omega_R & -2i\Omega_R & 0 \end{pmatrix}, \tag{35}$$

$$M_{31} = \begin{pmatrix} 0 & 0 & i\alpha\Omega & 0 & 0 \\ 0 & 0 & 0 & -i\alpha\Omega & -i\Omega \\ 0 & 0 & 0 & 0 & i\alpha\Omega \end{pmatrix}, \tag{36}$$

$$M_{32} = \begin{pmatrix} i\Omega & 0 & 0 & 0 & i\Omega_R/2 \\ 0 & 0 & 0 & 0 & -i\Omega_R/2 \\ -i\alpha\Omega & 2i\Omega & -2i\Omega & -2/\tau_\sigma & 0 \end{pmatrix}, \tag{37}$$

$$M_{33} = \begin{pmatrix} -1/(\tau_a + \tau_\sigma) & 0 & i\Omega_R \\ 0 & -1/(\tau_a + \tau_\sigma) & -i\Omega_R \\ i\Omega_R & -i\Omega_R & -2/(\tau_a + \tau_\sigma) \end{pmatrix}. \tag{38}$$

The dimension of matrix (30) is 13 × 13; M_{11}, M_{12}, M_{21}, and M_{22} are 5 × 5 matrices; M_{33} is a 3 × 3 matrix; M_{13} and M_{23} are 5 × 3 matrices; and M_{31} and M_{32} are 3 × 5 matrices.

Equation set (27) describes the dynamics of the interacting atom and nanoparticle in the field of an external wave. The set is linear, which means that the spectrum of an atom in the presence of a nanoparticle can be determined using the quantum regression theorem.

We recall that the observation of a triplet spectrum requires that the condition $|\Omega| = |\mathbf{E}\boldsymbol{\mu}_{\mathrm{TLS}}|/\hbar \gg \tau_\sigma^{-1}$ be valid. At the same time, the restriction imposed on the magnitude of the external field by the small-plasmon-number condition is $\Omega \ll \tau_a^{-1}$. The experimental decay constant for metallic nanoparticles is $\tau_a \sim 10^{-14}$ s and that for semiconductor quantum-dot implementations of the two-level system is $\tau_\sigma \sim 10^{-1}$ s, i.e., $\tau_\sigma^{-1} \ll \Omega \ll \tau_a^{-1}$. Thus, there is a range of external-field amplitudes ($\tau_\sigma^{-1} \ll \Omega \ll \tau_a^{-1}$) where the atom spectrum has a triplet shape, while the number of plasmons in the nanoparticle is small. Thus, in a wide range of external fields, the effect of the nanoparticle on the resonance fluorescence spectrum may be analyzed under the assumption that the number of plasmons is small.

4. ATOM SPECTRUM IN THE PRESENCE OF THE NANOPARTICLE

According to the Wiener–Khinchin theorem, the spectrum $S(\omega)$ is given by the Fourier transform of the two-time correlation function $\langle E(t+\tau)E^{+}(t)\rangle$. We assume hereinafter that the dipole moments of the nanoparticle and the two-level atom are collinear and oriented parallel to the x axis, the electromagnetic wave is linearly polarized along the x axis and propagates along the y axis, and the observation point is located on the z axis. Then, $E < \langle(\mathbf{r}, t+\tau)E^{+}(\mathbf{r}, t)\rangle = I_0(\mathbf{r})\langle[\alpha\hat{a}^{+}(t+\tau) + \hat{\sigma}^{+}(t+\tau)][\alpha\hat{a}(t) + \hat{\sigma}(t)]\rangle$, where $I_0(\mathbf{r}) = (\omega_{\mathrm{TLS}}^{2}|\boldsymbol{\mu}_{\mathrm{TLS}}|/c^2|\mathbf{r}|)^2$. The factor $I_0(\mathbf{r})$ will be dropped in the calculations below. Thus, in order to calculate the spectrum of the system, we need to

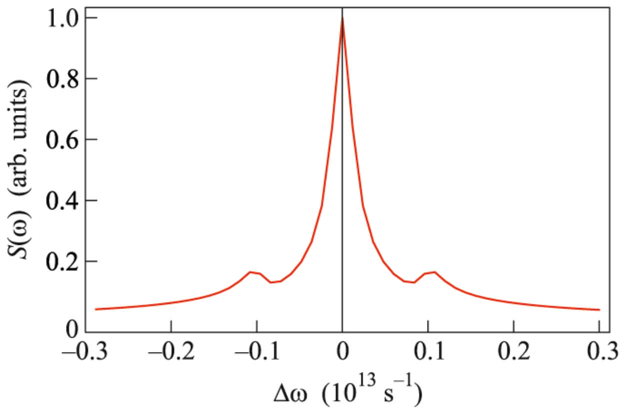

Fig. 1. Spectrum of the atom–nanoparticle system in an external filed for $\Omega_R = 0$.

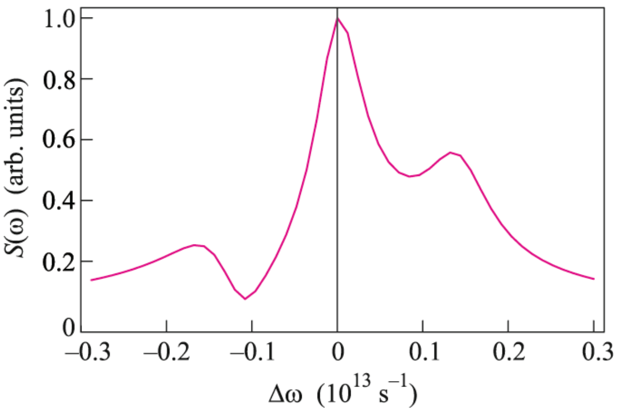

Fig. 2. Same as in Fig. 1, but for $\Omega_R = 4 \times 10^{12}$ s^{-1}.

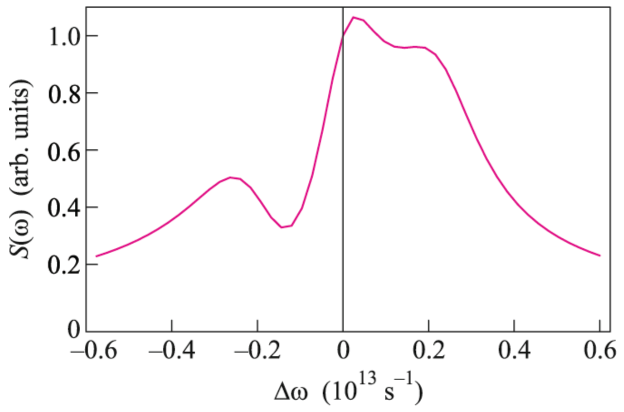

Fig. 3. Same as in Fig. 1, but for $\Omega_R = 8 \times 10^{12}$ s^{-1}.

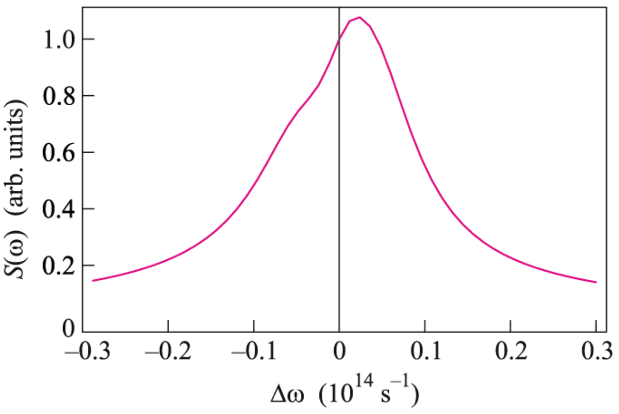

Fig. 4. Same as in Fig. 1, but for $\Omega_R = 2 \times 10^{13}$ s^{-1}.

find the operator averages $\langle \hat{a}^+(t+\tau)\hat{a}(t)\rangle$, $\langle \hat{a}^+(t+\tau)\hat{\sigma}(t)\rangle$, $\langle \hat{\sigma}^+(t+\tau)\hat{a}(t)\rangle$, and $\langle \hat{\sigma}^+(t+\tau)\hat{\sigma}(t)\rangle$. These quantities can be determined by applying the quantum regression theorem, expressed by Eq. (6), to equation set (27). In this way, the following set of equations for the required average values is obtained:

$$\frac{\partial}{\partial \tau}\langle \hat{x}_i(t+\tau)\hat{x}_j(t)\rangle = M_{ik}\langle \hat{x}_k(t+\tau)\hat{x}_j(t)\rangle + x_{0i}\langle \hat{x}_j(t)\rangle, \tag{39}$$

here, $\hat{\mathbf{x}}$, $\mathbf{x}_0$, and M are given by Eqs. (28)–(30), respectively.

Equation set (39) was solved numerically. The Fourier transforms of $\langle E(\mathbf{r}, t+\tau)E^+(\mathbf{r}, t)\rangle$ are shown in Figs. 1–4. In the case where the nanoparticle and the atom do not interact (Fig. 1), the spectrum represents a sum of the resonance fluorescence triplet (Mollow triplet) and the broad line of the nanoparticle. As the nanoparticle–atom interaction increases, the side peaks of the resonance fluorescence spectrum merge into the widening central peak. Characteristically, this "merger" occurs asymmetrically for each peak. First, the right peak gets "swallowed" (Fig. 2), so that only two peaks (the central and left ones) remain (Fig. 3). One can see that the spectrum assumes the shape of a Fano resonance line, typical of situations with two interacting oscillators. In the case under study, the external field acts to excite the system, and a shift between the phases of the dipole moments sets in only owing to the interaction between the nanoparticle and the atom.

As the strength of the interaction between the nanoparticle and the atom increases further, the left spectral peak becomes absorbed as well. When this interaction is much stronger than the interaction with the external field (i.e., $\Omega_R \gg \Omega$), the spectrum represents a Lorentzian line whose width is proportional to Ω_R^2, i.e., to the square of the atom–nanoparticle coupling constant (Fig. 4).

5. CONCLUSIONS

Thus, we have investigated the effect of the interaction between a nanoparticle and an atom on the resonance fluorescence spectrum. We have shown that the shape of the Mollow triplet is modified upon an increase in the atom–nanoparticle interaction strength. First, the right peak disappears and the spectrum assumes a shape typical of a Fano resonance line. The physical origin of this behavior can be understood if we consider the interacting nanoparticle and two-level atom as two coupled oscillators. Owing to their interaction, the phase shift between the oscillating dipole moments of the nanoparticle and the atom depends on the detuning between the external excitation frequency and the oscillator resonant frequencies. The resulting interference leads to a Fano resonance feature superimposed on the resonance-fluorescence spectrum. An important fact is that, for a certain strength of the atom–nanoparticle interaction, the right peak of the Mollow triplet disappears, while the left peak is still present. As the interaction becomes still stronger, the spectrum degenerates into a Lorentzian line whose width is proportional to the square of the atom–nanoparticle coupling constant Ω_R^2.

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