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18

Modeling of resonance fluorescence excitation spectra of two different two-level interacting nanoparticles

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Numerical simulations of the excitation spectra of resonant fluorescence bound by the dynamic interaction of two different two-level nanoparticles have been performed. We considered the excitation of one of the particles by monochromatic light with a frequency close to the transition frequency of this particle, at an irradiation intensity leading to a small population of its excited state. The formalism for the description of resonance fluorescence by the Heitler–Ma method was applied to a complete compound system of the marked particle pair and quantized emission field. The resulting expressions for the excitation spectrum reflect consideration of the absorption of the fluorescence photon without attributing it to the emission of any one particle. Expressions for the excitation spectra when taking into account such absorption of the fluorescence photon that allows us to attribute it to the radiation of one or another of the particles were obtained by solving a modified system of equations of the Heitler–Ma method, which reflects the addition of non-interfering amplitudes of the emission field states of each particle. It was found that the shape of the excitation spectra of the stationary resonance fluorescence of the particles in question is determined by the shape of the corresponding intensity spectra of the spontaneous fluorescence of these particles.

Keywords: mathematical modeling, interacting two-level nanoparticles, resonant fluorescence, excitation spectrum.

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Introduction

The results of modeling the dynamics of states of two-level nanoparticles irradiated by monochromatic light and bound by dynamic interaction are often used to interpret experimental data on various aspects of resonant fluorescence of two isolated atoms, ions, and semiconductor quantum dots (see, for example, [1] and the literature cited there). In many works, the noted modeling is based on the application of the semi-classical formalism of Bloch optical equations for a closed system of particles in the basis of their collective states. A much more complete modeling based on the formalism of the theory of an open, composite system of particles and quantized radiation field, whose states are subjected to external influence („measurement"). This formalism was used in [2,3] to simulate the population dynamics and spectral characteristics of two dynamically coupled closely spaced different two-level particles and their spontaneous fluorescence (SF). These works used the notion of dividing the Hilbert space of states of a composite system into subspaces, which were used to describe the SF, which ends with the photon absorption by the macroscopic environment of the particles when their collective ground state is populated, or when any one of the particles populates the ground state.

Based on the use of a similar formalism, this paper simulates the type of excitation spectra of the stationary RF of the two particles noted above. We considered the excitation of one of the particles by a long pulse of resonant monochromatic irradiation, which does not lead to a significant settlement of its excited state.

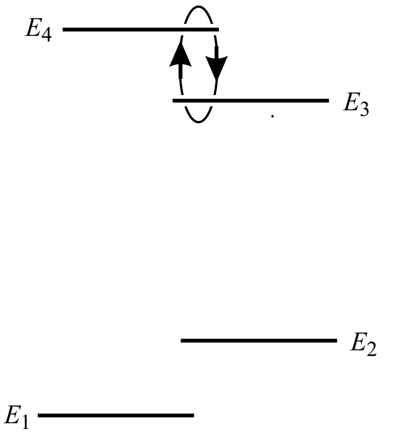
Model of a complete composite system

We write the Hamiltonian of a complete composite system of two „point", stationary two-level particles (particle A and particle B), which interact with each other and with the quantized radiation field, as

$$H = H_f + W + \sum_{j=A,B} {}_jH + {}_jV,$$

where H_f — the Hamiltonian of the radiation field; W — the dynamic interaction energy operator between particles;${}_AH({}_BH)$ — the Hamiltonian of particle $A(B)$; ${}_AV = -(\mathbf{e}\mathbf{d}_A)$ — the electrodipole energy operator ($\mathbf{d}_A$ — the dipole moment operator of particle A) of particle A interaction with transverse radiation field components and the similar expression ${}_BV = -(\mathbf{e}\mathbf{d}_B)$ for particle B (λ — the set of indices, characterizing the wave vector of the photon and its polarization state). The orientation of the particle transition dipoles and the direction of irradiation are such that it excites only particle A. The distance between the particles is much smaller than the wavelength of free particle A and the wavelength of free particle B.

Let's introduce the designations: $|n\rangle(n = 1, 4)$ — eigenstates of particle A with energies E_1 and E_4; $|n\rangle(n = 2, 3)$ —



Schematic of the relative arrangement of particle energy levels with indication of non-radiational transitions between them.

eigenstates of particle B with energies E_2 and E_3; $\hbar\omega_{nm} = (E_n - E_m)$, where $n = 4, 3, 2$; $m = 3, 2, 1$; $n > m$; $\omega_{41} - \omega_{32} = \omega_{43} + \omega_{21} \equiv 2\hbar\Delta$, where $0 < \Delta \ll \omega_{41}, \omega_{32}$.

The figure schematically shows the relative positions of the horizontal lines marked by horizontal energy levels with approximately equal values of the difference $E_4 - E_3 > 0$ and $E_2 - E_1 > 0$ (as is often taken when considering the simulation of interacting atom ion state dynamics — see, for example, Fig. 1, c in [1]). Note that in many papers on the state dynamics of two-level particles — see, for example, [4] and the literature cited there — it has been assumed that $E_2 = E_1$. The circular curves in this figure denote non-radiational, reversible transitions between particles in excited states. They are conditioned by the action of the considered W operator, by which we will mean the dipole-dipole interaction energy operator of particles, as, for example, adopted in describing the interaction of two closely spaced alkaline earth atoms whose fluorescence intensity beat was studied in [5], or the CdSe/ZnS quantum dot pair, whose relative position geometry was determined by studying their RF in [6].

The $|\lambda\rangle$ state is taken as the initial state of the irradiation field. It contains an integer number $N > 1$ of photons λ with the frequency ω_L from the area of frequencies satisfying the condition $\omega_{41} \gg |\omega_L - \omega_{41}|$; λ — the set of indices that describe the photon wave vector and its polarization state. We denote the field state containing $N - 1$ photons λ as $|0\rangle$.

We will take into account the possibility of different types of registration of the fluorescence photon state $\mu(\mu \neq \lambda)$ at its continuous absorption by the environment or a special detector (photon counter). We denote a field state that contains $N - 1$ photons λ and a photon μ whose absorption populates the collective ground state of the particle pair as $|\mu\rangle$ and call the corresponding RF the „non-selective"one. The field state containing $N - 1$ photons λ and a fluorescence photon whose absorption populates the state $|1\rangle$ („discarding all others" — see page 46 in [7]), we denote as$|\sigma\rangle$ $(\sigma \notin \mu)$, and for such absorption, which is accompanied by a state $|2|rangle$, we denote it as $|\nu\rangle$ $(\nu \notin \mu, \sigma \neq \mu)$. We will call the RF for such registrations of its photon states as the „selective RF"one. Remarks on the treatment of the physical meaning of separating of subspaces of states $|\sigma\rangle$ and $|\nu\rangle$ from the common space of states $||mu\rangle$ see, for example, [2].

For the volume L^3 of space in which there are particles and the considered radiation field, we will denote the matrix element of the operator ${}_AV$ by the states of the complete composite system $|12\lambda\rangle$ and $|420\rangle$ as follows:

$$\langle 12\lambda|{}_AV|420\rangle = iL^{-3/2}\sqrt{2\pi N\hbar\omega_L}(d_\lambda)_{14} \equiv V^{40}_{1\lambda} = i\hbar\Omega,$$

where $(d_\lambda)_{14}$ — the matrix element on the states $|1\rangle$ and $|4\rangle$ projection of the operator d on the direction of photon polarization λ, Ω — „frequency of Rabi". Hereinafter, for simplicity, the indices $V^{420}_{12\lambda}$ and $V^{420}_{12\sigma}$ are omitted, as well as the indices „2"in $V^{130}_{12\mu}$ and $V^{130}_{12\nu}$ — indices „1". The integer is $N > 1$, but it is small enough so that the population of the state $|4\rangle$ is much less than 1. For the matrix element of the operator ${}_AV$ by states $|12\mu\rangle$ and $|420\rangle$, we use this designation:

$$\langle 12\mu|{}_AV|420\rangle = iL^{-3/2}\sqrt{2\pi\hbar\omega_\mu}(d_\mu)_{14} \equiv V^{40}_{1\mu},$$

where $(d_\mu)_{14}$ — the matrix element on the states $|1\rangle$ and $|4\rangle$ of the operator d projection on the polarization direction of photon μ, ω_μ — the photon frequency μ. Similarly, we use the designations:

$$\langle 12\mu|{}_BV|130\rangle = iL^{-3/2}\sqrt{2\pi\hbar\omega_\mu}(d_\mu)_{23} \equiv V^{30}_{2\mu},$$

$$\langle 12\sigma|{}_AV|420\rangle = iL^{-3/2}\sqrt{2\pi\hbar\omega_\sigma}(d_\sigma)_{14} \equiv V^{40}_{1\sigma}$$

and

$$\langle 12\nu|{}_BV|130\rangle = iL^{-3/2}\sqrt{2\pi\hbar\omega_\nu}(d_\nu)_{23} \equiv V^{30}_{2\nu}.$$

The matrix elements of the operator W between the states $|420\rangle$ and $|130\rangle$ will be denoted as $\langle 42|W|13\rangle = \langle 13|W|42\rangle \equiv \hbar w$.

The simulation of the population dynamics of the states of a complete compound system will be based on the use of solutions of the system of Schr?dinger equations for the amplitudes $b_k(t)$ of the population of the eigenstates of the accepted Hamiltonian in the interaction representation:

$$i\hbar\dot{b}_k(t) = \sum_i ({}_AV + {}_BV + W)_{kl} b_l(t)$$
$$\times \exp\big[i(E_k - E_l)t/\hbar\big] + i\hbar\delta_{ki}\delta(t),$$

where i, k, l — sets of indices denoting the basis ortho-normalized states of the total system. The energies of these states are designated respectively as E_i, E_k, E_l; i — the aggregate indices of the initial state $|12\lambda\rangle$; k and l — the aggregate indices of „intermediate" states $|420\rangle$, $|130\rangle$ or „final"states $|12\mu\rangle$ of non-selective RF or „final" states $|12\sigma\rangle$, $|12\nu\rangle$ of selective RF; δ_{ki} — the Kronecker symbol, $\delta_{ki} = 0$ at $k \neq i$ and $\delta_{ki} = 1$ at $k = i$.

The use of the above matrix elements of the interaction operator of particles and the radiation field corresponds to a description of the dynamics of the states of a complete compound system that takes into account the transitions with conservation of its energy, neglecting the small contribution to the amplitude of these states from accounting for virtual transitions, at which the energy of the complete system is not conserved („resonance approximation“). This description of the state dynamics of the complete composite system reflects the conservation of stability of the $|12\sigma\rangle$, $|12\nu\rangle$ states of the considered subspaces.

Time is counted from the moment $t = 0$. Regarding the role of the nonhomogeneous term $i\hbar\delta_{ki}\delta(t)$ and the preservation of the normalization $\sum_k |b_k(t)|^2 = 1$ — see, for example, §16 ch. 4 in [8]. When using Fourier representations

$$b_k(t) = i(2\pi)^{-1}\int\limits_{-\infty}^{+\infty} G_{ki}(E)\exp[i(E_k - E)t/\hbar]dE,$$

$$i\hbar\delta(t) = i(2\pi)^{-1}\int\limits_{-\infty}^{+\infty} \exp[i(E_i - E)t/\hbar]dE$$

matrix $G_{ki}(E)$ is determined by solving a system of equations:

$$(E - E_k)G_{ki}(E) = \sum_l ({}_AV + {}_BV + W)_{kl}G_{li}(E) + \delta_{ki}.$$

In work [9], it was proposed to represent the matrix $G_{ki}(E)$ as

$$G_{ki}(E) = U_{ki}(E)G_{ii}(E)\xi(E - E_k),\ \ k \neq i,\ \ U_{ii} \equiv 0,$$

where $\xi(E) = P/E - i\pi\delta(E)$, P/E — the principal value of the function $1/E$, the matrix $U_{ki}(E)$ is defined by the system of equations (16.9)−(16.12) given in [8]. This approach to finding a solution to the Schr?dinger equation in the interaction representation is commonly referred to as the „method of Heitler–Ma“.

As applied to the aforementioned description of the non-selective RF, the system of equations of the Heitler–Ma method (without specifying here and below, for simplicity, the initial state index in the elements U_{ki} and Γ_{ii}) in the resonance approximation has the form

$$U_{420}(E) = V^{1\lambda}_{40} + \hbar w U_{130}(E)\xi(E - E_{130})$$
$$+ \sum_\mu V^{1\mu}_{40}U_{12\mu}(E)\xi(E - E_{12\mu}),$$

$$U_{130}(E) = \hbar w U_{420}(E)\xi(E - E_{420})$$
$$+ \sum_\mu V^{2\mu}_{30}U_{12\mu}(E)\xi(E - E_{12\mu}),$$

$$U_{12\mu}(E) = V^{40}_{1\mu}U_{420}(E)\xi(E - E_{420})$$
$$+ V^{30}_{2\mu}U_{130}(E)\xi(E - E_{130}),$$

$$\hbar\Gamma(E) = iV^{40}_{1\lambda}U_{420}(E)\xi(E - E_{420}).$$

The two-component equation for $U_{12\mu}(E)$ reflects the summation of the „interfering alternatives“ (see page 25 in [10]) photon emission probabilities of particle A and particle B.

As noted above, the consideration of selective RF is based on the partitioning of the state space $|12\mu\rangle$ into a state subspace $|12\sigma\rangle$ and a state subspace $|12\nu\rangle$. Instead of the element $U_{12\mu}(E)$ we will consider two elements: $U_{12\sigma}(E)$ and $U_{12\nu}(E)$. Accordingly, instead of the equation for $U_{12\mu}(E)$ we have the equation for $U_{12\sigma}(E)$ and the equation for $U_{12\nu}(E)$;in the equation for $U_{420}(E)$, the summation over μ will be replaced by the summation over σ, and in the equation for $U_{130}(E)$ — by the summation over ν. We will denote these elements as $\breve{U}_{420}(E)$ and $\breve{U}_{130}(E)$. Taking the above assumptions and designations, the system of equations for the selective RF has the form

$$\breve{U}_{420}(E) = V^{1\lambda}_{40} + \hbar w\breve{U}_{130}(E)\xi(E - E_{130})$$
$$+ \sum_\sigma V^{1\sigma}_{40}U_{12\sigma}(E)\xi(E - E_{12\sigma}),$$

$$\breve{U}_{130}(E) = \hbar w\breve{U}_{420}(E)\xi(E - E_{420})$$
$$+ \sum_\nu V^{2\nu}_{30}U_{12\nu}(E)\xi(E - E_{12\nu}),$$

$$U_{12\sigma}(E) = V^{40}_{1\sigma}\breve{U}_{420}(E)\xi(E - E_{420}),$$

$$U_{12\nu}(E) = V^{30}_{2\nu}\breve{U}_{130}(E)\xi(E - E_{130}),$$

$$\hbar\breve{\Gamma}(E) = iV^{40}_{1\lambda}\breve{U}_{420}(E)\xi(E - E_{420}).$$

This system of equations reflects a description of the dynamics of the „incompatible alternatives “ (see p. 26 in [10]) of the probability of a photon RF being emitted by either particle A or particle B (an effect of „the photon emission path “ — see ch. 20 in [11]).

Solving equations and type of excitation spectra

When solving the above systems of equations we will use the designations

$$i\hbar\gamma_4(E) = -\sum_\sigma |V^{40}_{1\sigma}|^2\xi(E - E_{12\sigma}),$$

$$i\hbar\gamma_3(E) = -\sum_\nu |V^{30}_{2\nu}|^2\xi(E - E_{12\nu}),$$

$$i\hbar\gamma_{(34)}(E) = -\sum_\mu V^{40}_{1\mu}V^{2\mu}_{30}\xi(E - E_{12\mu}).$$

We will not take into account radiative shifts of particle energy levels, leaving in these sums only the terms proportional to $\delta(E)$. Given that the photon frequency of the stationary RF coincides with the irradiation frequency (see footnote on p. 231 in [8]), we imply by the remaining expressions $\gamma_m (m \div 4, 3)$ and $\gamma_{(34)}$ the usual constants γ_4, γ_3 and $\gamma_{(34)}$ which characterize the radiation decay rates of the $|4\rangle$ state of the isolated particle A, respectively the $|3\rangle$ state of the isolated particle B and the joint radiation decay of these states. Using these designations, we obtain the solution to the system of equations for the non-selective RF

$$U_{420}(E) = V_{40}^{1\lambda}(E - E_{420})\tilde{E}_i(E)/F_{(\mu)}(E),$$

$$U_{130}(E) = V_{40}^{1\lambda}(E - E_{130})\hbar(w - i\gamma_{(34)})/F_{(\mu)}(E),$$

$$U_{12\mu}(E) = V_{40}^{1\mu}\left[V_{1\mu}^{40}\tilde{E}_I(E) + V_{2\mu}^{30}\hbar(w - io\gamma_{(34)})\right]/F_{(\mu)}(E),$$

$$\Gamma_{(\mu)}(E) = i\hbar\Omega^2\tilde{E}_I(E)/F_{(\mu)}(E),$$

where

$$\tilde{E}_I(E) \equiv E - E_{130} + i\hbar\gamma_3,$$

$$F_{(\mu)}(E) \equiv \left[\tilde{E}_I\tilde{E}_{II} - \hbar^2(w - i\gamma_{(34)})^2\right],$$

$$\tilde{E}_{II}(E) \equiv E - E_{420} + i\hbar\gamma_4.$$

Similarly, we obtain the solution of the system of equations for the selective RF

$$\breve{U}_{420}(E) = V_{40}^{1\lambda}(E - E_{420})\tilde{E}_I(E)/F(E),$$

$$\breve{U}_{130}(E) = V_{40}^{1\lambda}(E - E_{130})\hbar w/F(E),$$

$$U_{12\sigma}(E) = V_{40}^{1\lambda}V_{1\sigma}^{40}\tilde{E}_I(E)/F(E),$$

$$U_{12\nu}(E) = V_{40}^{1\lambda}V_{2\nu}^{30}\hbar w/F(E),$$

$$\breve{\Gamma}(E) = i\hbar\Omega^2\tilde{E}_I(E)/F(E),$$

where $F(E) \equiv (\tilde{E}_I\tilde{E}_{II} - \hbar^2 w^2)$.

For the light transmutation of the considered complete composite system, the expression $\mathrm{Re}[\Gamma(E_{12\lambda})]$ determines the „total probability of RF per unit time“ (p. 234 in [8]). We will call the dependence of $\mathrm{Re}[\Gamma(E_{12\lambda})]$ on ω_L the „spectrum of the excitation“ of the considered stationary RF during photon λ conversion, noting that this term often refers to the dependence of intensity („power“ by [11]) of RF on the frequency of monochromatic irradiation of the light scattering particle.

Applying the accepted definition of the RF excitation spectrum, denoting it as $S_F(\bar{\omega}_L)$ and using the resulting solution $\Gamma_{(\mu)}(E)$, we find

$$\mathrm{Re}[\Gamma(E_{12\lambda})] \equiv S_F(\bar{\omega}_L)$$

$$= \Omega^2\Big\{\gamma_4[\bar{\omega}_L + 2\Delta)^2 + \gamma_3^2] + \gamma_3(w^2 + \gamma_{(34)}^2)$$

$$+ 2\gamma_{(34)}[w(\bar{\omega}_L + 2\Delta) - \gamma_3\gamma_{(34)}]\Big\}/\Phi_{(\mu)}(\bar{\omega}_L),$$

where

$$\Phi_{(\mu)}(\bar{\omega}_L) = \hbar^{-4}|F_{(\mu)}(\bar{\omega}_L)|^2 = (\bar{\omega}_L^2 + \gamma_4^2)[(\bar{\omega}_L + 2\Delta)^2 + \gamma_3^2]$$

$$+ 2(\gamma_{(34)}^2 - w^2)\left[\bar{\omega}_L(\bar{\omega}_L + 2\Delta) + \gamma_3\gamma_4\right]$$

$$- 4w\gamma_{(34)}[\gamma_4(\bar{\omega}_L + 2\Delta) + \gamma_3\bar{\omega}_L] + (w^2 + \gamma_{(34)}^2)^2.$$

If $\gamma_{(34)} = 0$, then we have an excitation spectrum $S_F(\bar{\omega}_L, \gamma_{(34)} = 0) \equiv S_{fAB}(\bar{\omega}_L)$ stationary unselective RF with such photon absorption μ, which allows „in principle“ (see note on p 459 in [11]) to attribute it either to a photon emitted by particle A or to a photon emitted by particle B, „without any selection of them“ (p. 54 in [7]). The excitation spectrum $S_{fAB}(\bar{\omega}_L)$ has the following form:

$$S_{fAB}(\bar{\omega}_L) = \mathrm{Re}[\breve{\Gamma}(E_{12\lambda})]$$

$$= \Omega^2\left\{\gamma_4[(\bar{\omega}_L + 2\Delta)^2 + \gamma_3^2] + \gamma_3 w^2\right\}/\Phi_S(\bar{\omega}_L),$$

where

$$\Phi_S(\bar{\omega}_L) \equiv \hbar^{-4}|F(\bar{\omega}_L)|^2 = (\bar{\omega}_L^2 + \gamma_4^2)[(\bar{\omega}_L + 2\Delta)^2 + \gamma_3^2]$$

$$- 2\bar{\omega}_L w^2(\bar{\omega}_L + 2\Delta) - 2\gamma_3\gamma_4 w^2 + w^4.$$

According to the formalism used, under the above conditions, there is equality

$$\frac{d}{dt}|b_k(t \to \infty)|^2 = 2\pi\hbar^{-1}|U_{ki}(E_k)|^2\delta(E_k - E_i),$$

so that $|U_{12\sigma}|(E_{12\sigma})|^2$ defines the excitation spectrum of the stationary RF emitted by particle A, i.e., corresponds to the registration of the RF photon emitted by this particular particle with corresponding information about the population fraction of its ground state in the population of collective ground state of this pair of particles. Let us denote the excitation spectrum of such an RF as $S_{fA}(\bar{\omega}_L)$. Using the solution $U_{12\sigma}|(E_{12\sigma})$, after summing $|U_{12\sigma}|(E_{12\sigma})|^2$ for all directions of photon σ propagation, considering energy conservation during transitions between states of the complete compound system, we find

$$S_{fA}(\bar{\omega}_L) = \Omega^2\gamma_4[(\bar{\omega}_L + 2\Delta)^2 + \gamma_3^2]/\Phi_S(\bar{\omega}_L).$$

Accordingly, $|U_{12\nu}|(E_{12\nu})|^2$ defines the excitation spectrum $A_{fB}(\bar{\omega}_L)$ of the stationary RF particle B and similarly to the above, we find

$$S_{fB}(\bar{\omega}_L) = \Omega^2\gamma_3 w^2/\Phi_S(\bar{\omega}_L),$$

so $S_{fA}(\bar{\omega}_L) + S_{fB}(\bar{\omega}_L) = S_{fAB}(\bar{\omega}_L)$.

It is easy to see that the obtained expressions for the components of the RF excitation spectra contour differ from the expressions for the corresponding components of the SF intensity spectra contour given in [3] only by the common multipliers. Therefore, Figs. 2 and 3 of [3], which show the appearance of the SF intensity spectra for the considered pair of particles, can be seen as an illustration that the appearance of the contours of the stationary RF excitation spectra is similar to that of two mutually overlapping SF intensity spectrum contours. Note that the appearance of the contours of the spectra $S_{fA}(\bar{\omega}_L)$ and $S_{fB}(\bar{\omega}_L)$ is similar to the appearance of two mutually overlapping contours of the steady-state oscillation spectrum of two different harmonic damped oscillators, one of which is excited by harmonic force (the mechanical analog of Fano contours — see, for example, in [12]).

Note that the particle pair considered in [2] was meant to be the simplest model of the reaction center of a bimolecular secondary photoreaction accompanied by SF. As a result of [2] modeling the dynamics of such photoreaction and comparing its results with the corresponding experimental data known from the literature, it was concluded that under normal conditions, such reactions selectively populate the ground states of the reagent molecules and the product molecules, i.e., the observed SF is selective. There is reason to believe that the stationary RF in bimolecular photoreactions is also selective, and the spectra $S_{fA}(\bar{\omega}_L)$ and $S_{fB}(\bar{\omega}_L)$ can be considered as the excitation spectra of the reagent molecule and the product molecule of such reactions. The dependences of $S_{fA}(\bar{\omega}_L)$ and $S_{fB}(\bar{\omega}_L)$ on the irradiation parameters and the particle structure parameters can be used to select such values of these parameters, which correspond to the creation of the optimal mode of the corresponding bimolecular photoreaction.

Closing remark

Based on the use of the Heitler-Ma formalism for studying RF, expressions determining the shape of the two-component contours of the excitation spectra of nonselective and selective stationary RF pairs of interacting two-level nanoparticles were obtained. It was found that the shape of these contours coincides with the shape of the contours of the corresponding spectra of the SF intensity of the nanoparticles in question.

Conflict of interest

The author declares that he has no conflict of interest.

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