PAPER

Self-organised aggregation of a pair of particles with different resonant frequencies and electric dipole moments of transitions, controlled by an external quasi-resonant field*

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Self-organised aggregation of a pair of particles with different resonant frequencies and electric dipole moments of transitions, controlled by an external quasi-resonant field*

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Abstract. **The influence of the oscillation phases of the dipole moments induced in metal nanoparticles and quantum dots by an external laser field on their interaction energy is considered. It is shown that a difference in resonant frequencies leads to the formation of additional minima and maxima, which are absent in the spectral dependence of the interaction energy of identical particles at similar orientations of the pair of particles with respect to the plane of polarisation of radiation. These features are due to the fact that the oscillation phase difference of the induced dipole moments of particles reaches values close to π.**

Keywords: *interaction of laser radiation with matter, dipole interaction, nanoparticles, nanostructures.*

1. Introduction

In recent years, the interest of researchers in the formation of nanostructures with certain specified properties constantly increases. Particular attention is paid to self-organisation processes, which are widespread in living nature and allow it to form very complex functional structures without any technological procedures. Currently, some success has been made in this field, which is promising for nanoscience [1–3]. However, self-organisation methods do not always allow one to obtain structures with desired and reproducible properties.

One of the ways to solve this problem is to use external physical effects under which controlled self-organisation of particles occurs. A method of forming structures with specified geometry from metal nanoparticles with resonances in the optical range using laser irradiation was proposed in [4]. Laser irradiation of microscopic objects leads to their polarisation, due to which particles can interact with both the external field (and thus form structures with sizes of the same order as the radiation wavelength [5–8]) and the other particles. In the latter case structures with sizes much smaller than the wavelength of the laser radiation inducing this polarisation can be formed. The particle optical resonances enhance the particle–particle interaction and serve as a basis for selective formation of various structures with specified arrangement of particles in aggregate.

In most studies devoted to the interaction of particles in an electromagnetic field, the objects of study were atoms with transition frequencies close to the laser frequency [9–11]. However, the attention of researchers is now focused on the interaction of nanoparticles with both the field and other particles. For example, the quantum effects caused by the interaction of the quadrupole moments induced on a neutral particle with an external electric field were considered in [12]. The formation of narrow collective plasmon resonances in the transmission spectra of one- and two-dimensional arrays of metal nanoparticles under their dipole interaction was comprehensively investigated in [13].

Our previous study [14] was devoted to the analysis (in the dipole–dipole approximation) of the interaction between metal and semiconductor nanoparticles (quantum dots) with differing resonant frequencies and widths of absorption lines. In contrast to a pair of metal particles [4], additional minima were revealed in the spectral dependence of the interaction energy. These minima can be related to the phase relations between the oscillations of induced dipole moments of individual particles.

In this paper, we report the results of studying the influence of the oscillation phases of the dipole moments of paired particles, induced by an external field, on their interaction energy in an external light field.

2. Basic calculation relations

The processes of self-organised aggregation of particles in a field of laser radiation can be described using the following physical model [4, 14]. If particles are located in a laser radiation field and are in thermodynamic equilibrium with the environment, the distances between some particles may become rather small due to their Brownian random walks, and the interaction between the oscillating dipole moments induced on these particles begins to significantly affect the character of their motion. In this situation the energy of electrodynamic interaction of the particles may exceed the thermal motion energy kT and the energy of the barrier impeding uncontrolled aggregation; as a result, particles may aggregate and form nanostructures in a configuration specified by the external field.

Let us consider an ensemble composed of N particles located at points with coordinates r_i and interacting with each other via polarisation forces induced by an external light field. The consideration will be performed in the dipole–dipole approximation. We assume the ensemble to be much less in

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size than the incident radiation wavelength; hence, the external field $\boldsymbol{E}$ can be considered as homogeneous for the entire ensemble and the delay effects can be neglected. Then the vector of the electric component of the light wave field can be written as a harmonic function dependent on only time:

$$\boldsymbol{E} = \tfrac{1}{2}\boldsymbol{E}_0 \exp(\mathrm{i}\omega t) + \text{c.c.}$$

In the vicinity of each particle the contributions of all other particles to the local field should be taken into account by summing the fields of the dipoles induced on all particles. In this case, the vector of a dipole moment with a frequency ω, induced on the ith particle, has the form

$$\boldsymbol{d}_i = \chi_{0i}\left(\boldsymbol{E} + \sum_{j\neq i}\boldsymbol{E}_j\right), \tag{1}$$

where $\chi_{0i} = |d_{12i}|^2/[\hbar(\Omega_i + \mathrm{i}\Gamma_i)]$ is the linear polarisability of an isolated particle in the two-level scheme under consideration; $|d_{12i}|^2$ is the squared modulus of the electric dipole moment of the transition of the ith particle between states 1 and 2; $\Omega_i = \omega_{0i} - \omega$ is the detuning from resonance; $\hbar$ is Planck's constant; Γ_i is the homogeneous linewidth;

$$\boldsymbol{E}_j = \frac{3(\boldsymbol{d}_j\boldsymbol{r}_{ij})\boldsymbol{r}_{ij} - \boldsymbol{d}_j\boldsymbol{r}_{ij}^2}{r_{ij}^5} \tag{2}$$

is the field formed by the jth particle of the ensemble at the point of location of the ith particle [15, 16]; and $r_{ij} = r_i - r_j$ is the distance between the ith and jth particles.

The interaction energy of the ith and jth dipoles is

$$W_{d_{ij}} = \frac{(\boldsymbol{d}_i\boldsymbol{d}_j)r_{ij}^2 - 3(\boldsymbol{d}_i\boldsymbol{r}_{ij})(\boldsymbol{d}_j\boldsymbol{r}_{ij})}{r_{ij}^5}. \tag{3}$$

Thus, one can calculate the interaction energy between particles by solving a system of N equations with respect to the dipole moments of all particles, which are determined by expression (1).

Let us consider two very simple cases: (i) interaction between silver and semiconductor particles with different frequencies and transition widths and (ii) interaction between two silver nanoparticles with coinciding transition frequencies. Let the external light field make an angle α with the straight line that connects the centres of two particles and is oriented parallel to the x axis. According to (1) and (2), we obtain a system of equations for determining the projections of the dipole moments d_{ix} and d_{iy} of the particles on the x and y axes:

$$\begin{aligned} d_{1x} &= \chi_{01}\left(E\cos\alpha + \frac{2d_{2x}}{r_{12}^3}\right), \quad d_{1y} = \chi_{01}\left(E\sin\alpha - \frac{2d_{2y}}{r_{12}^3}\right), \\ d_{2x} &= \chi_{02}\left(E\cos\alpha + \frac{2d_{1x}}{r_{12}^3}\right), \quad d_{2y} = \chi_{02}\left(E\sin\alpha - \frac{2d_{1y}}{r_{12}^3}\right), \end{aligned} \tag{4}$$

where r_{12} is the distance between the particle centres.

The solutions to system (4), with allowance for the dependence of the linear polarisability of an isolated particle on the squared modulus of the electric dipole moment of transition, detuning from resonance, and the absorption linewidth, have the form

$$\begin{aligned} d_{1x} &= r_{12}^3 E\omega_{s1}\cos\alpha \\ &\times\left[\frac{(\Omega_2 + 2\omega_{s2})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) + \Gamma_2(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right. \\ &+\left.\mathrm{i}\frac{\Gamma_2(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) - (\Omega_2 + 2\omega_{s2})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right] \\ &\times\exp(\mathrm{i}\omega t) + \text{c.c.}, \\ d_{1y} &= r_{12}^3 E\omega_{s1}\sin\alpha \\ &\times\left[\frac{(\Omega_2 - \omega_{s2})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) + \Gamma_2(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right. \\ &+\left.\mathrm{i}\frac{\Gamma_2(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) - (\Omega_2 - \omega_{s2})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right] \\ &\times\exp(\mathrm{i}\omega t) + \text{c.c.}, \\ d_{2x} &= r_{12}^3 E\omega_{s2}\cos\alpha \\ &\times\left[\frac{(\Omega_1 + 2\omega_{s1})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) + \Gamma_1(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right. \\ &+\left.\mathrm{i}\frac{\Gamma_1(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) - (\Omega_1 + 2\omega_{s1})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right] \\ &\times\exp(\mathrm{i}\omega t) + \text{c.c.}, \\ d_{2y} &= r_{12}^3 E\omega_{s2}\sin\alpha \\ &\times\left[\frac{(\Omega_1 - \omega_{s1})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) + \Gamma_1(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right. \\ &+\left.\mathrm{i}\frac{\Gamma_1(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) - (\Omega_1 - \omega_{s1})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2})^2 + (\Omega_1\Gamma_2 + \Omega_2\Gamma_1)^2}\right] \\ &\times\exp(\mathrm{i}\omega t) + \text{c.c.}, \end{aligned} \tag{5}$$

where $\omega_{si} = |d_{12i}|^2/(\hbar r_{12}^3)$ is the frequency shift of the resonance of the ith particle due to its interaction with a neighbouring one.

It is convenient to write the expression for the energy of dipole–dipole interaction between particles in a form containing the amplitudes and phases of the induced dipole moments of all particles. Then, after averaging over time, expression (3) takes the form

$$W_d = \frac{|d_{1y}||d_{2y}|\cos(\varphi_{1y} - \varphi_{2y})\sin^2\alpha - 3|d_{1x}||d_{2x}|\cos(\varphi_{1x} - \varphi_{2x})\cos^2\alpha}{4r_{12}^3}, \tag{6}$$

where $|d_{1y}|$, $|d_{2y}|$ and $|d_{1x}|$, $|d_{2x}|$ are the moduli and φ_{1y}, φ_{2y} and φ_{1x}, φ_{2x} are the phases of the y and x components of the time-averaged dipole moments of the first and second particles, respectively.

Using formula (5), one can write the phases of the dipole moments as

$$\varphi_{1x} = \arctan\frac{\Gamma_2(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) - (\Omega_2 + 2\omega_{s2})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_2 + 2\omega_{s2})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) + \Gamma_2(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)},$$

$$\varphi_{2x} = \arctan\frac{\Gamma_1(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) - (\Omega_1 + 2\omega_{s1})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1 + 2\omega_{s1})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - 4\omega_{s1}\omega_{s2}) + \Gamma_1(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}, \quad (7)$$

$$\varphi_{1y} = \arctan\frac{\Gamma_2(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) - (\Omega_2 - \omega_{s2})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_2 - \omega_{s2})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) + \Gamma_2(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)},$$

$$\varphi_{2y} = \arctan\frac{\Gamma_1(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) - (\Omega_1 - \omega_{s1})(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}{(\Omega_1 - \omega_{s1})(\Omega_1\Omega_2 - \Gamma_1\Gamma_2 - \omega_{s1}\omega_{s2}) + \Gamma_1(\Omega_1\Gamma_2 + \Omega_2\Gamma_1)}.$$

Further calculations will be performed with the following particle parameters. Silver nanoparticles with a radius r = 6 nm have maximum absorption (resonance) at λ_r = 420 nm, homogeneous line FWHM $\Delta\lambda$ = 90 nm, and squared modulus of the electric dipole moment of the transition $|d_{12}|^2 = 3.12 \times 10^{-30}$ erg cm^3 [14, 16]. The corresponding values for a CdSe quantum dot, according to [17, 18], are as follows: r = 5 nm, λ_r = 620 nm, $\Delta\lambda$ = 3 nm, and $|d_{12}|^2 = 1.91 \times 10^{-31}$ erg cm^3.

3. Calculation results

The case of interaction between two identical metal particles, which was described in [4], is similar to the case of two interacting permanent dipole moments (Fig. 1).

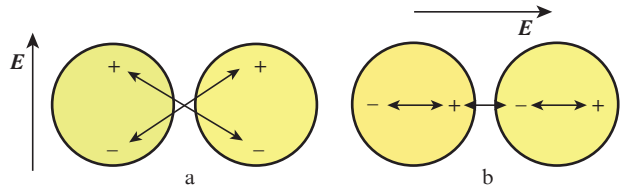

Figure 1. Electrical interaction of induced dipoles for a field directed (a) orthogonally and (b) collinearly with respect to pairs of particles.

According to expression (7), the oscillation phase difference for a pair of identical particles is zero for all external field wavelengths. Then, if the plane of polarisation of external field is parallel to the line connecting the particle centres (Fig. 1b) (i.e., the angle α = 0), the first term in expression (6) is zero and the second term is negative. This indicates formation of a potential well in the interaction energy spectrum and, as a consequence, attraction between the particles. At the same time, if the plane of polarisation is perpendicular to the line connecting the particles (i.e., the angle α = 90°), the second term in (6) is zero and the first term is positive. In this case, a maximum arises in the interaction energy spectrum, and particles are repulsed (Fig. 1a). These considerations are valid only when the cosine of the phase difference remains positive for all external field wavelengths. For a pair of metal particles [4] the phase difference of dipole moment oscillations is zero for all wavelengths.

If the transition dipole moments, linewidths, or particle resonant frequencies are different, the oscillation phases of the dipole moments will differ, in correspondence with (7). Therefore, there may be a situation where the phase difference is nonzero and exceeds $\pi/2$; in this case, additional minima or maxima are formed in the interaction energy spectrum [14].

Let us consider two very simple cases: (i) interaction of two identical metal particles and (ii) interaction between metal and semiconductor particles, with the aforementioned parameters and the distance r_{12} between the particle centres equal to the sum of particle radii.

The calculation scheme corresponds to the mutual arrangement of the particles and orientation of the plane of polarisation as in Fig. 2a. Figures 2b and 2c present the calculated dependences of the dipole–dipole interaction energy of pairs of particles (normalised to the thermal energy) on the angle α and the external field wavelength. We chose the following values of the parameters entering the equation: external field E = 200 esu and temperature T = 300 K.

The data of Figs 2b and 2c indicate that the interaction between particles with different resonant frequencies, linewidths, and transition dipole moments gives rise to additional minima and maxima in the interaction energy spectrum, which are due to the nonzero phase difference of dipole moment oscillations. At α = 0, the occurrence of a maximum in the case of interacting metal and semiconductor particles,

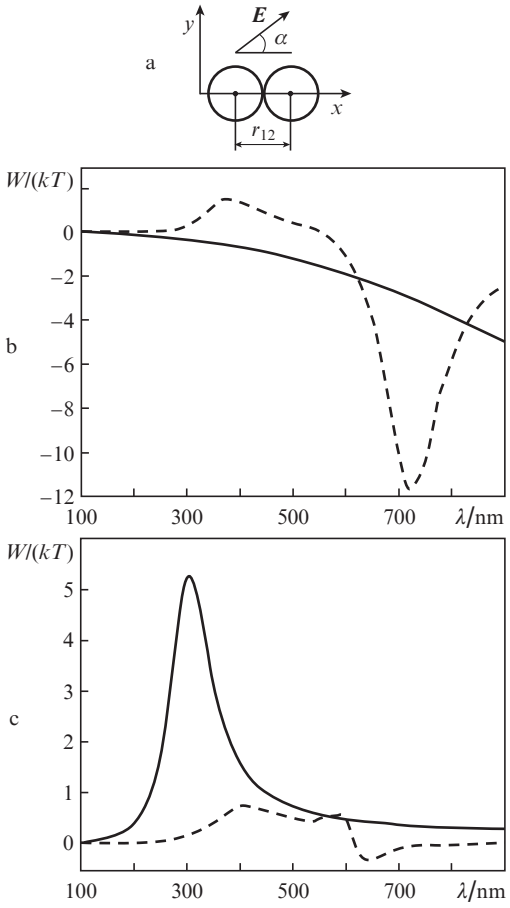

Figure 2. (a) Calculation scheme and (b, c) the spectral dependences of the dipole–dipole interaction energy for (solid lines) two metal particles and (dashed lines) metal and semiconductor particles at angles α = (b) 0 and (c) 90°.

is due to the phase shift of dipole-moment oscillations, when the cosine of the phase difference is negative in the wavelength range from 300 to 400 nm. Thus, the second term in expression (6) is positive, and the particles are repulsed. At $\alpha = 90°$ a minimum arises near 630 nm, which corresponds to attraction of particles; this feature is atypical of such mutual orientation of a pair of metal particles and the plane of polarisation of the external field.

As follows from the above expressions, the interaction between particles leads to a shift of their resonant frequencies. In addition, this interaction can also affect the phase difference of dipole moment oscillations. For example, we will analyse the behaviour of the oscillation phases of the dipole moments in a pair of semiconductor particles with differing radii in two cases: disregarding and taking into account their interaction. The resonant wavelength [17] and the dipole moment of the transition of semiconductor particles are known to depend on the particle radius. Figure 3 shows the calculated oscillation phases for the x component of polarisation in the cases of semiconductor particles with radii of 2.9 nm (solid line) and 5.4 nm (dashed line), with resonant wavelengths of 508 and 768 nm, respectively.

Figure 3a presents the known behaviour of the oscillation phases of the dipole moments of isolated particles to illustrate the effect of interaction. With allowance for the interaction between particles with different resonant frequencies and transition dipole moments (Fig. 3b), the specific features of the oscillation phase behaviour in the wavelength range of 440–580 nm give rise to an additional maximum in the interaction energy spectrum. This maximum is formed because the phase difference reaches π and the dipole moments begin to oscillate in antiphase; according to Fig. 1a, particles are repulsed in this case.

A similar situation is observed for the oscillation phases of the y component of dipole moments (Fig. 4), where the phase difference reaches π at $\lambda \sim 810$ nm, due to which a potential well is formed.

Thus, a difference in the particle resonant frequencies induces repulsion forces for the x component of the dipole moments and attraction forces for their y component; this is not characteristic of the stationary interaction of dipole moments and particles with coinciding resonances.

Let us consider the influence of the difference in the resonant wavelengths and electric dipole moments of the transition of semiconductor particles on the depth of the potential well formed and on the phase difference of dipole-moment oscillations for the y component. These values are known to be related to quantum dot sizes. The dependence of the resonant wavelength on the particle radius can be approximated based on the experimental data of [17], and the dependence of the squared modulus of the transition dipole moment on the resonant wavelength λ_0 has the form

$$|d_{12}|^2 = \frac{3h\sigma c\lambda_0}{4\pi\Delta\lambda}, \tag{8}$$

where $\sigma = 2.7 \times 10^{-19}$ cm^2 is the absorption cross section [18].

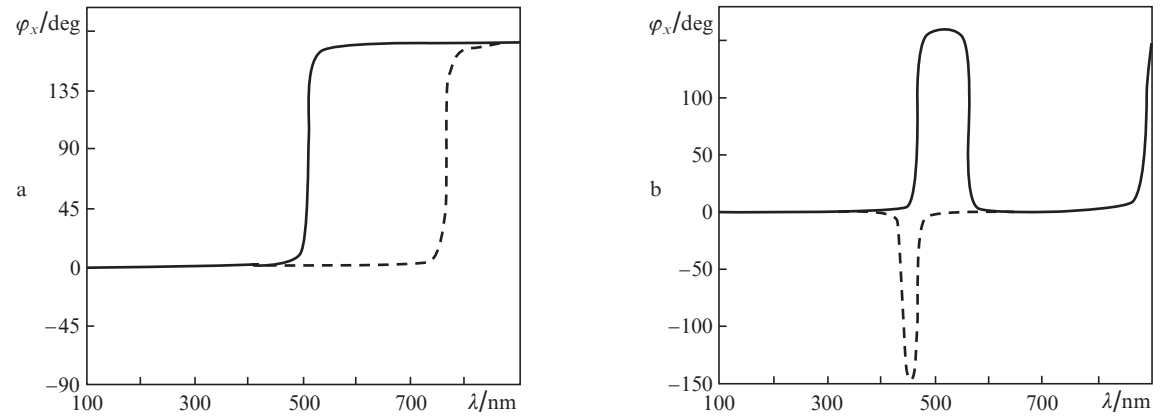

Figure 3. Phases of dipole-moment oscillations for the x component of polarisation in the cases of two (a) noninteracting and (b) interacting semiconductor particles.

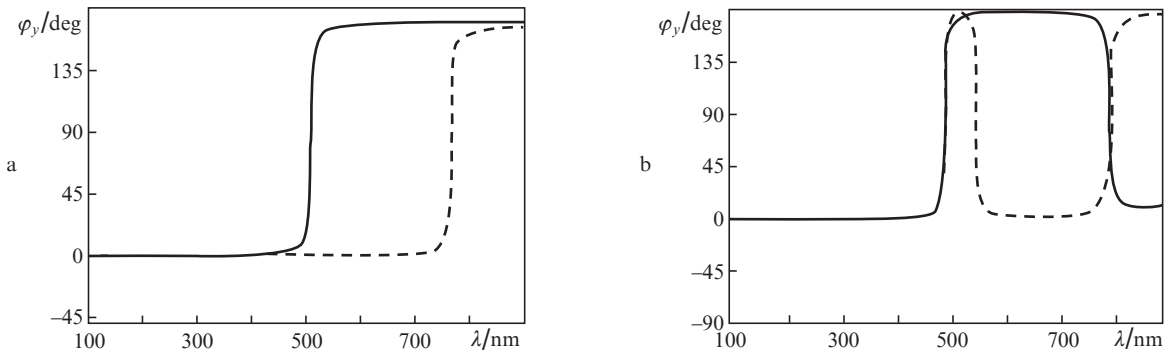

Figure 4. Same as in Fig. 3 but for the y component of polarisation.

Figure 5 shows the dependence of the potential-well depth on the wavelength difference for the y component of the field, which is due to the phase difference of the oscillations induced by dipole moments. The resonant wavelength remains fixed for the first particle but changes for the second one.

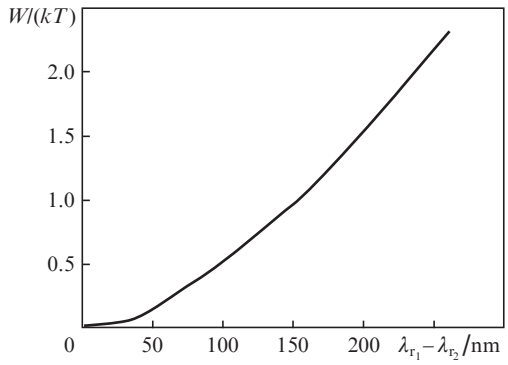

Figure 5. Dependence of the potential-well depth on the difference in the particle resonant wavelengths $\lambda_{r_1} - \lambda_{r_2}$.

As can be seen in Fig. 5, the potential-well depth increases with increasing difference in the resonant wavelengths and transition dipole moments. These data are interpreted in Fig. 6, which shows the dependence of the phase difference on the resonant-wavelength difference.

According to Fig. 6 and expression (6), the difference in the resonant wavelengths, linewidths, and electric dipole moments of transitions leads, first, to a change in the sign of the cosine of the phase difference in (6) and occurrence of attraction between particles. Second, for the y component, the first term in (6) increases, because the cosine of the phase difference approaches unity. Note that the phase difference changes from zero to values close to π at a difference in the resonant wavelengths exceeding the resonant linewidth $\Delta\lambda$ by approximately an order of magnitude.

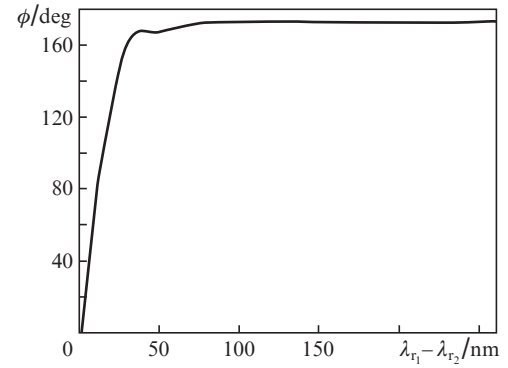

Figure 6. Dependence of the phase difference ϕ of dipole-moment oscillations on the difference in the particle resonant wavelengths $\lambda_{r_1} - \lambda_{r_2}$.

4. Conclusions

We considered the fundamental difference of the interaction of the dipole moments induced by an external resonant field in particles characterised by different resonant wavelengths, electric dipole moments of transitions, and linewidths from the interaction between the dipole moments of identical particles and particles with a permanent dipole moment. The interaction between nonidentical nanoparticles not only shifts the resonances of isolated particles but leads also to the formation of specific features in the spectrum of phase oscillation difference for the induced dipole moments. These features are responsible for the additional minima and maxima in the interaction energy spectrum.

It is shown that the formation of a potential well at an angle $\alpha = 90°$ is due to the phase shift of the oscillations of induced dipole moments of particles by a value close to π.

An increase in the difference in the particle resonant wavelengths increases the depth of the potential well arising in the spectral dependence of the interaction energy. The oscillation phase difference, depending on the difference in the particle resonant wavelengths, rapidly increases from zero to a value close to π and then varies only slightly.

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