LIGHT ABSORPTION BY RANDOM CLUSTERS OF SILVER PARTICLES

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We present some results of theoretical and experimental studies of light absorption by random fractal clusters of quasispherical silver particles 5 *nm in radius. Aggregation of particles in clusters is found to be accompanied by the appearance of a long–wave wing in the absorption spectra, while the absorption at the frequency corresponding to absorption by a monomer decreases. The results of calculation of the wing shape by a technique of bonded dipoles describe fairly well the experimental data. The absorption band broadening is nonuniform, what is well confirmed by the experiments on burning out of spectral and polarization–selective dips in the absorption spectra of clusters.*

INTRODUCTION

In the process of association of solid particles that diffuse through gas or liquid, clusters can be formed from a lot of primary monomer particles. Prolonged process of aggregation results in the formation of fractal structures.[1] Studying the optical properties of cluster structures is of interest for various scientific fields such as physics of the atmosphere and water media, creation of nonlinear optical and recording media, analysis of trace impurities, and catalysis.

The clusters of noble–metal particles are very convenient medium for investigation, because the techniques of preparation of samples are well developed, the material constants are known and the problem of oxidizing does not become pressing. In experiments with clusters of noble–metal particles a number of interesting effects were elicited. Thus the effect of giant Raman scattering of light is well known.[2] In the process of aggregation of colloid silver particles in clusters a considerable increase in the efficiency of degenerate four–photon scattering was observed.[3]. Selective threshold photo–modification of clusters of silver particles was discovered in Ref. 4. Irradiation of clusters of silver particles by a high–power laser pulse results in burning out a dip in their absorption spectrum near the laser radiation wavelength. This dip is primarily observed for radiation having the same polarization as the linear laser one. The appearance of the dip correlates with changes in the cluster structure.[5]

Aggregation is accompanied by the large broadening of the absorption spectrum of the colloid solution. This fact was repeated noted (see Ref. 2), but the satistactory theoretical description of the spectrum was lacking.

In this paper we develop the procedure for calculation of the absorption spectrum of clusters and compare the results of calculation with the data obtained in experiment carried out with the silver hydrosoles.

As a rule, the existing natural clusters consist of different particles of very complex shape (the exception is some types of the so–called regular clusters, for example, consisting of ice crystals or snowflakes). Theoretical description of the optical properties of such clusters is a serious problem. There exists, however, a simple physical model that enables one to describe qualitatively and quantitatively the cluster properties observed in experiment. This model is based on the assumption that the cluster consists of uniform particles. In spite of very abstract character, this assumption is sufficiently universal. For example, when cluster consists of highly prolate particles, each of these particles can be represented smaller spherical particles. In general, as shown in Refs. 6 and 7, the optical properties of dielectric particle of arbitrary shape can be approximated well by an ensemble of smaller spherical particles embedded at sites in the cubic lattice and bounded by the surface of a primary particle.

In the theoretical section of this paper we accept in general a hypothesis that monomers are spherical and uniform. In addition we assume that radii of monomers are small compared with the wavelength of the exciting radiation. This assumption is valid for a great variety of clusters. The clusters of silver particles discussed in experimental section consist of monomers whose diameters are of the order of 10 nm while the wavelength in the spectral range area under investigation varies from 200 to 1200 nm. We note that the cluster dimensions as a whole may be rather large compared with the wavelength.

1. THEORY

1.1 Construction of a model. Let us consider the cluster that contains N spherical monomers with the permittivity ε placed in vacuum so that the permittivity $\varepsilon(\mathbf{r})$ is defined as

$$\varepsilon(\mathbf{r}) = \begin{cases} \varepsilon\,, & \mathbf{r} \in V_i\,,\ \ i = 1, \ldots, N\,, \\ 1\,, & \mathbf{r} \notin V_i\,, \end{cases} \qquad (1)$$

where V_i, is the volume occupied by the ith particle. The volume V_i is bounded by a sphere of radius a centered at the point $\mathbf{r}_i$. The geometry of cluster is completely determined by the vectors $(\mathbf{r}_1, \ldots, \mathbf{r}_N)$ for such a problem formulation. We denote by v the volume occupied by an individual monomer being equal to $4\pi a^3/3$. The interaction of a monochromatic electromagnetic field (temporal dependence of the form $\exp(-i\omega t)$ is further omitted) with a cluster is described by the wave equation

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$$\left[\Delta + k^2\,\varepsilon(\mathbf{r})\right] \mathbf{E}(\mathbf{r}) = 0\ , \tag{2}$$

where $\kappa = \omega / c$. By entering the vector of polarization **P(r)**, we rewrite Eq. (2) in the form

$$\left[\Delta + k^2\right] \mathbf{E}(\mathbf{r}) = -\,4\,\pi\,k^2\,\mathbf{P}(\mathbf{r})\ , \tag{3}$$

$$\mathbf{P}(\mathbf{r}) = \frac{\varepsilon(\mathbf{r}) - 1}{4\,\pi}\,\mathbf{E}(\mathbf{r})\ . \tag{4}$$

Let $\hat{G}(\mathrm{r},\ \mathrm{r}') / 4\pi k^2$ is the Green's dyad function of the system of equations (3). The factor $1/4\pi k^2$ is entered for convenience. Then instead of differential equation (3) we derive the integral equation of the following form:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{\mathrm{inc}}(\mathbf{r}) + \sum_{i=1}^{N} \int_{V_i} \hat{G}(\mathbf{r},\ \mathbf{r}')\,\mathbf{P}(\mathbf{r}')\,d\mathbf{r}'\ , \tag{5}$$

where the incident wave field $\mathbf{E}_{\mathrm{inc}}(\mathbf{r})$ is the solution of homogeneous wave equation (3). We assume that

$$\mathbf{E}_{\mathrm{inc}}(\mathbf{r}) = \mathbf{E}_0\,\exp\,(i\,\mathbf{k}\,\mathbf{r})\ , \tag{6}$$

which corresponds to excitation of a cluster by a plane monochromatic wave. The relation between **E(r)** and **P(r)** is given by expression (4) with dielectric function (1). We note that Eq. (5) is integrated only over the domains in which $\varepsilon(\mathbf{r}) = \varepsilon$.

The Green's function has simple physical sense. When a point dipole oscillating with the frequency ω and amplitude **d** is placed at the point **r**, the radiation field of this dipole at the point **r** will be $\mathbf{G}(\mathbf{r},\ \mathbf{r}',)\,\mathbf{d}$. Thus interpretation of integral equation (5) is that the total field at the point **r** is the super position of the external (exciting) radiation field and the field created by the dipole moments $\mathbf{P}(\mathbf{r}')\ d\mathbf{r}'$, distributed over the volumes V_i.

The integral equation (5) compared with Eq. (3) has simpler physical sense, which is why it is more convenient for analysis. In addition, it contains the volumes V_i as the domains of integration in an explicit form. This enables one to use this equation for derivation of approximate solutions based on sphericity of these volumes.

Let us write down the explicit expression for $\hat{G}$. The action of this operator on the arbitrary vector **d** is determined by the following formulas:

$$\hat{G}(\mathbf{r},\ \mathbf{r}') = \hat{G}(\mathbf{r} - \mathbf{r}'), \tag{7}$$

$$\hat{G}(\mathbf{R})\,\mathbf{d} = k^3\left[A(k\,R)\,\mathbf{d} + B(k\,R)\,\frac{(\mathbf{d}\ \mathbf{R})\,\mathbf{R}}{R^2}\right] - \frac{4\,\pi}{3}\,\delta(\mathbf{R}), \tag{8}$$

$$A(x) = \left[\frac{1}{x} + \frac{\mathrm{i}}{x^2} - \frac{1}{x^3}\right]\exp\,(\mathrm{i}\,x)\ , \tag{9}$$

$$B(x) = \left[-\,\frac{1}{x} + \frac{3\,\mathrm{i}}{x^2} - \frac{3}{x^3}\right]\exp\,(\mathrm{i}\,x)\ . \tag{10}$$

We note the following impotant property of the operator $\hat{G}$:

$$\int_{V_i} \hat{G}(\mathbf{r}_j,\ \mathbf{r}')\,d\mathbf{r}' = \begin{cases} \nu\,\hat{G}(\mathbf{r}_j,\ \mathbf{r}_i)\ , & i \neq j\ , \quad (11a) \\ -(4\,\pi/3)\,[3+2\,\exp\,(\mathrm{i}\,k\,a)\,(\mathrm{i}\,k\,a - 1)]\ , & i = j\ . \quad (11b) \end{cases}$$

Formula (11*b*) is the exact one for arbitrary relations between a and λ, where $\lambda = 2\pi / k$ is the wavelength. Taking into account that $a \ll \lambda$ ($k\,a \ll 1$), Eq. (11*b*) transforms into a relation.

$$\int_{V_i} \hat{G}(\mathbf{r}_i,\ \mathbf{r}')\,d\mathbf{r}' = -4\,\pi / 3\ . \tag{11c}$$

1.2. Derivation of the equation of bonded dipoles. In this section we use relations (11) to pass over from integral equation (5) to the system of linear algebraic equations in the dipole approximation.

The essence of the dipole approximation is the assumption of constancy of the polarization vector **P(r)** and field inside each volume V_i:

$$\mathbf{P}(\mathbf{r}) = \mathbf{P}_i,\ \mathbf{E}(\mathbf{r}) = 4\,\pi\,\mathbf{P}_i / (\varepsilon - 1),$$

$$\mathbf{E}_{\mathrm{inc}}(\mathbf{r}) = \mathbf{E}_0\,\exp\,(\mathrm{i}\,\mathbf{k}\,\mathbf{r}_i),\ \text{for } \mathbf{r} \in V_i\ . \tag{12}$$

The dipole approximation for an isolated particle is justified when the wavelength of the exciting radiation is much greater than the particle radius. Generally speaking, this condition is insufficient for the cluster. Really, the dimentions of field inhomogeneities created by the dipole radiation of particles may be comparable to their dimensions, even though the wavelenght of the external radiation is sufficiently large. The strong inhomogeneities of the field are created in the cluster only by the particles in close proximity (the nearest neighbors) for which the distance R in formula (8) describing the dipole radiation changes within the size of a particle from 0 (point of contact) to $4a$ (diametrically opposite points).

To overcome this difficulty, we applied the following method. The distance between the centers of two nearest particles was taken to be less than the double radius $2a$ of particles. In such a way, we allow for the fact that the field dimensions actually do not coincide with the particle size due to inhomogeneous polarization inside each particle. More specifically, the highest degree of polarization (in the volume of a particle) is reached close to the point of contact of this particle with its nearest neighbor rather than in its geometrical center. Usually [6,7] the relation between b and a is chosen starting from the condition of equality of the volume of a cubic cell with the edge b to the volume of a ball of radius a.

It then follows that

$$b / a = (4\,\pi / 3)^{1/3} \approx 1.612\ . \tag{13}$$

As shown in Refs. 6 and 7, the dipole approximation together with condition (13), for the fractal three–dimensional clusters (fractal dimensionality $D = 3$) furnishes correct results. One–dimensional linear monomers ($D = 1$) were studied by Markel′,[8] who demonstrated that in this case $b/a = 1.688$ must be chosen to provide the correct results. In intermediate cases ($1 < D < 3$) the values of b/a should probably be taken in the range from 1.612 to

1.688. Since the speed of b/a is very small, we will further use relation (13). It provides good agreement with experiment for clusters of silver particles ($D = 1.8$).

Substituting relation (12) in Eq. (5) and using Eqs. (11*a*) and (11*c*), we obtain the following linear system of equations for the dipole moments of monomers $\mathbf{d}_i = \mathrm{v}\,\mathbf{P}_i$:

$$\mathbf{d}_i = \chi\left[\mathbf{E}_0\,\mu_i + \sum_{i\neq j}\hat{G}(\mathbf{r}_i, \mathbf{r}_j)\,\mathbf{d}_j\right], \tag{14}$$

$$\chi = \frac{3\,\mathrm{v}}{4\,\pi}\,\frac{\varepsilon - 1}{\varepsilon + 2} = a^3\,\frac{e - 1}{\varepsilon + 2}, \tag{15}$$

$$\mu_i = \exp\left(\mathrm{i}\,\mathbf{k}\,\mathbf{r}_i\right). \tag{16}$$

It is seen from formula (15) that χ is the dipole susceptibility of a dielectric ball of radius a in the quasistatic approximation. With the use of more exact expression (11*b*) instead of Eq. (11*c*), the formula for χ assumes the following form:

$$\chi = a^3\,\frac{\varepsilon - 1}{\varepsilon + 2 + 2(\varepsilon - 1)\,[1 + (\mathrm{i}\,k\,a - 1)\,\exp\,(\mathrm{i}\,k\,a)]}. \tag{17}$$

Formula (17) is the next term in the Mie expansion of the dipole polarizability χ in the parameter ka. It allows for self–action of an oscillating dipole and so unlike formula (15) fits the optical theorem and obeys the law of conservation of energy.

Equation (14) is called the equation of bonded dipoles. Actually it is a $3N$–dimensional system of linear algebraic equations in Descartes' components of the vector of dipole moments $d_{i\alpha}$ with symmetrical complex matrix. The solution of this system is a set of the complex vectors $\mathbf{d}_1, \ldots, \mathbf{d}_N$ that can be used for calculating the characteristics of optical cluster.

1.3. Optical cross sections. The amplitude of scattering $\mathbf{f}(\mathbf{s})$ in the direction of arbitrary unit vector $\mathbf{s}$

$$\mathbf{f}(\mathbf{s}) = k^2 \sum_{i=1}^{N} [\mathbf{d}_i - (\mathbf{d}_i\,\mathbf{s})\,\mathbf{s}]\,\exp\,(-\mathrm{i}\,\mathbf{k}\,\mathbf{s}\,\mathbf{r}_i). \tag{18}$$

is of fundamental significance in calculation of the optical cross sections of extinction σ_e, scattering σ_s and absorption σ_a. The following relation

$$d\sigma_s / d\Omega = |\mathbf{f}(\mathbf{s})|^2, \tag{19}$$

holds for the differential scattering cross section, where $d\Omega$ is the element of the solid angle in the direction of scattering. The total scattering cross section can be derived by integration of Eq. (19), while the cross sections of extinction σ_e and absorption σ_a fit the optical theorem

$$\sigma_s = \int |\mathbf{f}(\mathbf{s})|^2\,d\Omega, \tag{20}$$

$$\sigma_e = \frac{4\,\pi}{k}\,\mathrm{Im}\,[\mathbf{f}(\mathbf{k}/\kappa)\,\mathbf{E}_0^*] / |\mathbf{E}_0|^2, \tag{21}$$

$$\sigma_a = \sigma_e - \sigma_s. \tag{22}$$

By integration of Eq. (20) with the use of formula (18) for $\mathbf{f}$ we derive the expression for the extinction and absorption cross sections

$$\sigma_e = \frac{4\,\pi\,k}{|\mathbf{E}_0|^2}\,\mathrm{Im}\sum_{i=1}^{N}\mu_i^*\,\mathbf{d}_i\,\mathbf{E}_0^*, \tag{23}$$

$$\sigma_a = \frac{4\,\pi\,k}{|\mathbf{E}_0|^2}\,[\mathrm{Im}\,(1/\chi^*) - 2\,k^3/3]\sum_{i=1}^{N}|\mathbf{d}_i|^2. \tag{24}$$

Formulas (20) and (21) and equation (14) provide a basis for the numerical calculation of the optical characteristics of clusters.

2. NUMERICAL SIMULATION

2.1 Simulation of the geometry of fractal clusters. To construct the model of random fractal clusters, we used the method of random wanderings without self–intersecting trajectories. The model yields the fractal dimensionality of cluster being equal to 1.78, which is close to the fractal dimensionality of the experimentally studied clusters of silver particles (see below). The experimentally measured radius a of monomers was 5 nm. The condition on the absence of self–intersecting trajectories was that the distances between any twomonomers may be no less than a certain value b chosen from formula (13). It then follows that $b = 1.612\ a = 8.06$ nm. If this condition was violated at the next step of random wanderings, it would be simply rejected. The step size in the method of random wanderings was also equal to b.

A totality of 25 random realizations of clusters were constructed, each contained $N = 30$ monomers. All solutions were further averaged over these random realizations.

2.2. Calculation of dipole susceptibility of a monomer. To calculate the dipole susceptibility of a monomer χ that enters equation of bonded dipoles (17), we used refined formula (17). It was assumed that the clusters were placed in water, as it was in the experiment. We borrowed the tabulated values of permittivity of silver ε (for bulk sample) from Ref. 9 and corrected then with due regard to the dimensionality effect that may be important for small metallic particles. The essence of this effect is that the conduction electrons are scattered at the boundary of a particle. We took into account the dimensionality effect in the following way. At first in the tabulated values of ε we separated the contribution of free electrons from the contribution corresponding to interzonal transitions. The former is described by the Drude formula

$$\varepsilon = 1 - \omega_p^2 / \omega\,(\omega + \mathrm{i}\,\Gamma). \tag{25}$$

Further we substitute $\mathbf{\Gamma} + \mathrm{v}_F / a$ for $\mathbf{\Gamma}$ where v_F is the Fermi velocity. Thereafter the corrected contribution of free electrons was added to the contribution corresponding to interzonal transitions to obtain the total permittivity. For silver we have $\lambda_p = 2\pi c / \omega_\mathrm{p} = 136.1$ nm, $\Gamma / \omega_\mathrm{p} = 0.0019$, and $\mathrm{v}_F / c = 0.0047$ (c is the light speed). With such values of the parameters for $a = 5$ nm the dimensionality effect results in the considerable change of permittivity. We note that dimensionality effect for scattering on isolated metallic particles was experimentally tested.[10]

2.3. Simulation of spectra and threshold photo–modification of clusters. The equation of the bonded dipoles was solved in two ways. The first square–root method is the direct exact method for inverting the matrix $\hat{G}$ of dipole interaction that effectively uses the symmetry of this matrix.

The second method of expansion in eigenvectors is iterative. It is applicable only to the real matrices $\hat{G}$ and enables one to shorten considerably the machine time.[11] The matrix $\hat{G}$ is real when the dimensions of a cluster as a whole are much smaller than the wavelength. In calculations by this method from formulas (9) and (10) we discarded the terms proportional to $1/x$ and $1/x^2$ (the contribution of the wave and intermediate zones) and assumed $\exp(ix) = 1$.

Both methods gave very close results though the dimensions of clusters under study were only slightly smaller than the wavelength. This confirms the conclusion of Ref. 12 that interaction clusters with the fractal dimensionality being less than 2 with the wave zone is insignificant.

We solved equation (12) for the wavelengths varying from 200 to 1400 nm. Further we used the obtained solutions to calculate the extinction cross sections according to formula (23). Efficiency of extinction Q_e shown in Fig. 1 as a function of λ is connected with it by the relation $Q_e = \sigma_e / N\pi a^2$.

We simulate the threshold photo–modification in the following way. The formula (24) shows that the energy absorbed by the ith monomer is proportional to $|\mathbf{d}_i|^2$. For non–interacting monomers $|\mathbf{d}_i|^2 = |\chi\mathbf{E}_0|^2 = \text{const}$. However, taking into account the effect of interaction, the value of $|\mathbf{d}_i|^2$ in a cluster under goes strong fluctuations and may considerably exceed $|\chi\mathbf{E}_0|^2$. The monomers that satisfy the condition $|\mathbf{d}_i|^2 / |\chi\mathbf{E}_0|^2 > h$ were removed from the cluster. (They were considered to be evaporated). We choose the value of h in such a way that about 5 clusters were removed from the totality of 25 clusters, which corresponded to a little excess of this value over the threshold.

After the monomers were removed, the cluster spectrum was calculated again on two different states of polarization: parallel and perpendicular to the polarization of the radiation burning out a dip. Thereafter the difference spectra (Fig. 1) were calculated. As can be seen from Fig. 1, photo–modification results in the appearance of spectral and polarization–selective dips in the spectrum of extinction.

3. EXPERIMENT

The silver hydrosoles were prepared by the boron–hydride method[13] and their diagnostics was carried out with an electron microscope. The newly-prepared colloid solution contained isolated silver elementary particles of nearly spherical shape 10–20 nm in diameter whose number density was $10^{12}\,\text{cm}^{-3}$. In the course of time the particles in the colloid aggregate in clusters consisting of tens or hundreds of monomers. A portion of monomers in our solution did not aggregate or weakly aggregate (clusters contained only a few monomers). Samples were stabilized by addition of gelatin in the solution. The absorption spectra were recorded with a spectrophotometer in a cavity 1 cm thick.

The results are shown in Fig. 2. The absorption spectrum of the isolated silver particles in water reaches a maximum near λ = 400 nm. The shape of the measured spectrum agrees fairly well with the results of calculation. In the process of aggregation particles in clusters, a long–wave wing occurs in the spectrum. The intensity and width of the wing increase for higher degree of aggregation. Figure 2 shows that the shape of the spectrum in the wing agrees fairly well with the results of calculation. At the same time the measured maximum value of absorption by the aggregated hydrosole of monomers is much higher than the calculated one. Apparently the excess absorption is due to the monomers which did not aggregate in the solution. When clusters are irradiated by laser pulses 5–10 ns long with energy being approximately equal to 10 mJ/cm² at the wavelengths corresponding to wing (λ_l = 532 , 576, and 1064 nm), the dip is burnt out in the absorption spectrum near λ_l (see curve *1* in Fig. 3) when the pulse has polarization the same as laser does. When the laser wavelength (λ_l = 437 nm) falls within the absorption maximum, the absorption on irradiation decreases everywhere over the region of the long–wave wing and increases in the region of maximum (see curve *2* in Fig. 3).

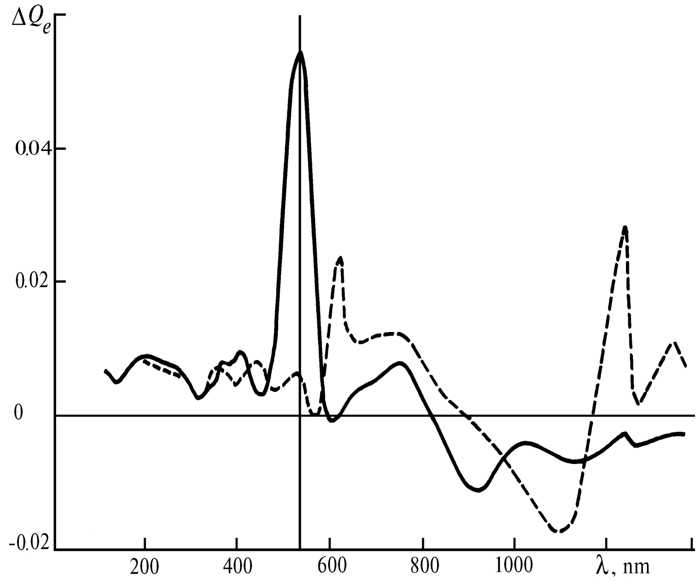

FIG. 1. Calculated difference spectra of clusters before and after irradiation by a high–power laser pulse at λ = 532 nm. The decrease of the absorption corresponds to the positive direction of the ordinate (spectral dip). Solid curve is for the same polarization as that of laser pulse ($\mathbf{e} \parallel \mathbf{e}_l$)*; and dashed curve is for the orthogonal polarization* ($\mathbf{e} \perp \mathbf{e}_l$).

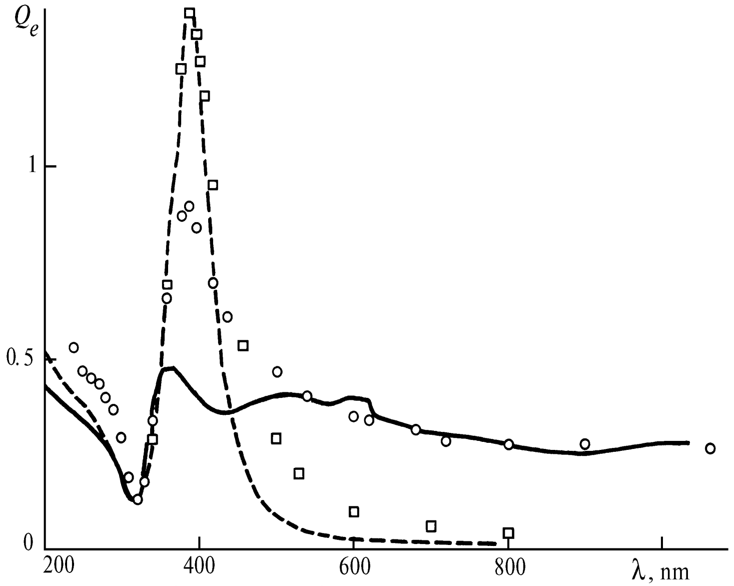

FIG. 2. *Theoretical spectra of monomers (dashed curve) and clusters consisting of 30 monomers (solid curve) compared with the experimental spectra of monomers (small squares) and clusters (small circles). The fractal dimensionality of clusters is close to 1.8.*

The results obtained can be interpreted in the following way. As follows from the foregoing theory, the wing in the spectrum occurs due to the dipole interaction of monomers in a cluster. The random of location of monomers results in the fact that different local configurations of monomers absorb radiation at different wavelengths. This means that the spectrum in the region of wing is nonuniformly broadened. When the radiation intensity exceeds a certain threshold value, the absorbing fragment of cluster is modified in such a way that it tunes away from resonance with the field. Accordingly, the dip occurs in the spectrum. The absorption at the frequency of maximum is cooperative in character.[11] The absorbed energy is smoothed out among monomers. A cluster dissociates upon exposure to radiation, and accordingly the absorption decreases everywhere over the region of the wing, while the maximum absorption increases.

The results of calculation of the spectrum of dips shown in Fig. 2 are in qualitative agreement with the experiment. The agreement is observed in the position of the dip and polarization characteristics. However, the experimentally measured dips are elongated in the long–wave region. Furthermore, the increase in the optical density of the irradiated sample is observed in the region around λ = 400nm in the measured difference spectrum, whereas the results of calculation show the increase in the density on the long–wave side of the dip. Apparently, a number of factors are operating to affect the experiment, which were neglected in the model. Among there factors are striction forces occurring in the cluster medium upon exposure to a powerful laser radiation field. The detailed discussion of this problem falls outside the scope of this paper.

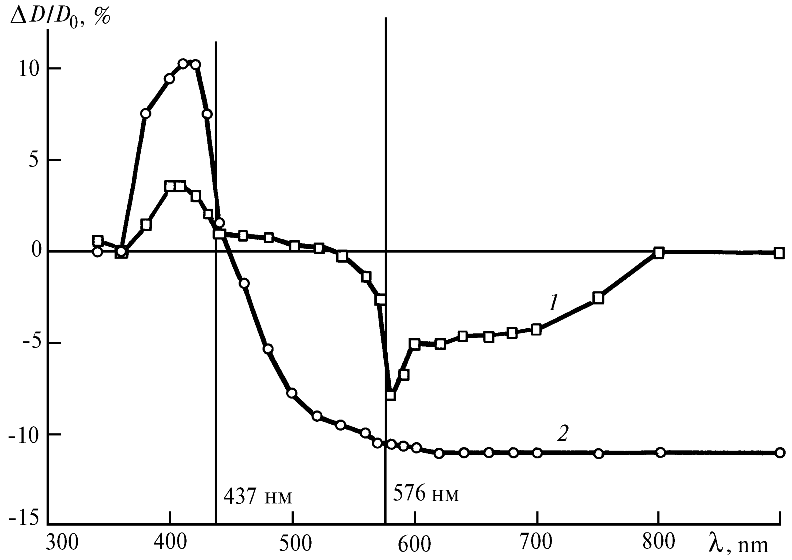

FIG. 3. *Difference absorption spectra of irradiated and non–irradiated cluster samples. The cavity is* 1 mm *thick. The laser radiation wavelength is* λ_l = 576 *(1) and* λ_l = 437 nm *(2).* ΔD *denotes a change in the optical density of sample, and* D_0 *denotes its optical density before irradiation.*

CONCLUSION

Both theoretically and experimentally we have shown that broad wings occur in the absorption spectra of random clusters consisting of loosely bound particles. The wings of the spectrum are nonuniformly broadened. The results of calculation of the spectrum of clusters of silver particles by the bonded dipole method indicate the presence of wing that agrees with the data of measurements. The theoretical conclusion about an important role of cooperative interaction of monomers as a result of absorption of radiation at the frequency of maximum is experimentally confirmed. The authors would like to express their gratitude to S.G. Rautian for helpful discussion of results and to A.I. Plekhanov for his assistance in the performance of the experiment.

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