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Optical extinction of an assembly of spherical particles in an absorbing medium: Application to silver clusters in absorbing organic materials

A.N. Lebedev[a] and O. Stenzel

Technische Universität Chemnitz, Institut für Physik, D-09107 Chemnitz, Germany

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Abstract. The theory presented by *Gerardy and Ausloos* for the calculation of the linear optical response of aggregates of spherical particles is analytically continued for absorbing embedding media. The method is based on the calculation of the extinction rate by a single particle embedded in an absorbing matrix. Explicit expressions for the extinction and scattering cross-sections are given. The method is applied to calculate the energy losses in several organic matrices with embedded silver clusters. Comparison with experimental data shows a very good agreement.

PACS. 78.20.Bh Theory, models, and numerical simulation – 78.66.Vs Small particles

1 Introduction

The interface region between metals and dielectrics is of particular interest in the field of modern applied optical spectroscopy. The possible excitation by optical means of propagating or localized plasmon modes in the metal fraction finds application in several surface-sensitive spectroscopic techniques, such as Surface Enhanced Raman Spectroscopy (SERS). Nowadays, these techniques are often applied to investigate the properties of organic molecular adsorbates on metal surfaces. Naturally, the extended π-electron-systems in these molecules may lead to significant absorption of the impinging light in the molecules. Thus the metal fraction (which may be a semi-space with a rough surface or simply an arrangement of metal clusters) is in touch with an *absorbing* environment here. Even when regarding the simplest case of a single spherical metal cluster in a homogeneous environment, the classical formulation of Mie's theory cannot be applied to the description of the optical properties of these systems, due to the absorption of the matrix.

Recently, we proposed a method to calculate the optical extinction rate of a single particle embedded in an absorbing medium [1]. Nevertheless, for real cluster matter, the single-particle assumption is certainly too crude, so that we have to generalize this formalism to the calculation of the optical response of coupled cluster aggregates. This is the aim of the present paper.

A general rigorous theory concerning optical properties of aggregated metal spheres was presented in 1981 by *Gerardy and Ausloos* [2]. It was shown that the effect of the electromagnetic interaction between the spheres may be important in obtaining the optical spectra of the system. The medium in which the clusters are immersed was supposed to be *non-absorbing*. The authors have mentioned that their method may also be applied for absorbing media introducing a complex wave vector in the matrix. Nevertheless, only a part of the problem can be solved in this way: one may find the electromagnetic fields in the matrix, but the meaning of the optical extinction by the aggregate in this case is not obvious and the explicit expressions for the extinction rate have to be found. An attempt to take into account absorption in the matrix was made also in [3] by considering each particle of the assembly to be coated with a shell of the matrix substance, which utilizes the solution of the problem of scattering on a coated sphere [4] and then implements the method of *Gerardy and Ausloos*. There are principal difficulties immediately seen if applying this model to real systems. First, the size of the shell may be empirically adjusted from the volume filling factor of the clusters in the matrix, but it can be done only in the case of uniformly distributed clusters. Second, the geometry of coated spheres involves different scattering processes if compared with the spheres immersed in a continuous absorbing host.

We will show that an explicit theoretical treatment of cluster aggregates in an absorbing medium is possible without the somewhat artificial assumption of aggregates of coated spheres.

The standard procedure to find the extinction and the scattering rate of the system of the particles in the case of a non-absorbing host matrix is to integrate the corresponding Poynting vectors over the surface of a large imaginary sphere, that encloses the entire assembly of the

[a] e-mail: a.lebedev@physik.tu-chemnitz.de

particles. The extinction is defined then as a sum of absorption and scattering rates given as the corresponding integrals over the sphere surface. However, the sphere construction is not possible for samples of limited size, for example, for thin films. Moreover, for absorbing embedding media this definition leads to an extinction value, that depends on the size of the integration volume, which obscures the physical meaning of the extinction in this case.

In our work [1] we have defined the extinction rate of the particle subsystem as the sum of absorption and scattering rates *in the particle volume* referenced to the matrix background. The rates were found by integrating the corresponding Poynting vectors *over the surface of the particle*. This provides a well-defined value for the particle extinction, that depends only on the *particle size* and the optical properties of the participating materials.

The main purpose of this work is the presentation of the method and we will briefly show how this method can be applied to calculate the optical energy losses in some cluster+absorbing matrix systems.

2 Theoretical

We consider an assembly of N spherical particles embedded in an absorbing matrix. The total electromagnetic field in the matrix is presented as a linear superposition of the incident (undisturbed by the presence of the particles) and the scattered fields. The incident light beam is assumed to be *already* in the matrix. Thus we omit all energy losses caused by the scattering from the two surfaces of the sample and neglect an interaction of the particles with the back-reflected light beams. We do not consider the possibility of optical excitation of longitudinal polarization waves both in the particle and in the matrix, so the wave field there is assumed to be transversal.

To find the scattered fields in the matrix, we follow closely the procedure suggested in [2], whose brief description is given here. The electromagnetic field incident on the surface of every single particle consists of the external incident wave and of the waves scattered by the other particles. Both the incident and the scattered fields and the fields in the particles are expanded in spherical coordinates in series of the spherical wave vector functions $\mathbf{m}_{kl}$ and $\mathbf{n}_{kl}$ (the definitions given in [2] are used). These expansions are written for every particle in the spherical coordinate system with the origin at the center of the particle. The attenuation of waves by the absorption in the matrix is totally described by introducing the complex wave vector $\mathbf{k}$ (as mentioned in [2]) of the field and using the complex argument kr in the definition of the spherical wave vector functions. Solving the ordinary boundary conditions on the surface of every i-th particle requires all field expansions to be transformed into the particles coordinate system and gives rise to the system of linear equations for the expansion coefficients of the scattered by the i-th particle field c^i_{qp} and d^i_{qp}:

$$c^i_{qp} = \Gamma^i_q \left\{ a^i_{0qp} + \sum_{l,m} \sum_{j \neq i} \left(c^j_{lm} \Im^{ij}_{lmqp} + d^j_{lm} \ell^{ij}_{lmqp} \right) \right\}, \quad (1)$$

$$d^i_{qp} = \Delta^i_q \left\{ b^i_{0qp} + \sum_{l,m} \sum_{j \neq i} \left(d^j_{lm} \Im^{ij}_{lmqp} + c^j_{lm} \ell^{ij}_{lmqp} \right) \right\}, \quad (2)$$

where a^i_{0qp} and b^i_{0qp} are the expansion coefficients of the external incident field in the vicinity of the i-th particle, Γ^i_q and Δ^i_q are its 2^q-polar magnetic and electrical susceptibilities, and $\Im^{ij}_{lmqp}$ and ℓ^{ij}_{lmqp} the elements of the transformation matrices for the spherical wave vector functions defined as

$$\Im^{ij}_{lmqp} = \left\langle \mathbf{m}^i_{lm3} | \mathbf{m}^j_{qp1} \right\rangle = \left\langle \mathbf{n}^i_{lm3} | \mathbf{n}^j_{qp1} \right\rangle , \quad (3)$$

$$\ell^{ij}_{lmqp} = \left\langle \mathbf{m}^i_{lm3} | \mathbf{n}^j_{qp1} \right\rangle = \left\langle \mathbf{n}^i_{lm3} | \mathbf{m}^j_{qp1} \right\rangle . \quad (4)$$

Here the functions $\mathbf{m}^i_{lm}$ and $\mathbf{n}^i_{lm}$ are defined in the coordinate system of the i-th particle and the subscripts 1 or 3 determine which of the spherical Bessel functions [5] $j_q(kr)$ or $h^{(1)}_q(kr)$ should be used in the radial part of the generating function for the spherical wave vector functions. The calculation of the elements of the transformation matrices is the main problem in finding the scattered fields in the matrix and is discussed in detail in [2]. The summations over $l \in [1, \infty)$ and $m \in [-l, l]$ in (1, 2) are truncated, so that $l \in [1, s]$, which provides an approximation to the coupling effect. The system (1, 2) is reformulated and solved in a matrix form

$$\mathbf{c} = \left[\left(\underline{\Gamma}^{-1} - \underline{\Im} \right) - \underline{\ell} \left(\underline{\Delta}^{-1} - \underline{\Im} \right)^{-1} \underline{\ell} \right]^{-1} \times \left[\mathbf{a}_0 + \underline{\ell} \left(\underline{\Delta}^{-1} - \underline{\Im} \right)^{-1} \mathbf{b}_0 \right] , \quad (5)$$

$$\mathbf{d} = \left(\underline{\Delta}^{-1} - \underline{\Im} \right)^{-1} \left[\mathbf{b}_0 + \underline{\ell} \mathbf{c} \right] , \quad (6)$$

where the matrices $\underline{\Gamma}$, $\underline{\Delta}$, $\underline{\ell}$, $\underline{\Im}$ have a dimension $Ns(s+2)$ for an assembly of N particles.

In our recent paper [1] we have defined the extinction rate of the particle subsystem W_e as the sum of absorption and scattering rates in the particle volume referenced to the matrix background. The absorption W_a and scattering W_s rates of the particle subsystem were found by integrating the corresponding Poynting vectors over the surface of the particle, leading to the following expression of the particle extinction:

$$W_\mathrm{e} = W_\mathrm{a} + W_\mathrm{s} - W_\mathrm{i} , \quad (7)$$

where W_i is an integral of the Poynting vector of the incident (non-disturbed by the presence of the particle) electromagnetic wave over the particle surface and gives the

absorption rate in the volume of the particle if filled with the matrix. In the case of the assembly of the particles we apply the same approach considering the entire aggregate as a "composite particle". So, an integration over the surface of such a "composite particle" is mathematically identical to an integration over the surface S_i of every i-th particle with the subsequent summation over all particles of the assembly. Such an integration of the Poynting vector of the total electromagnetic field gives the absorption rate in the assembly W_a

$$W_\mathrm{a} = -\frac{1}{2}\mathrm{Re}\sum_i \oint_{S_i} [\mathbf{E}_\mathrm{t} \times \mathbf{H}_\mathrm{t}^*] \cdot \overrightarrow{\mathrm{d}s}\,, \tag{8}$$

where $\overrightarrow{\mathrm{d}s}$ is in the direction of an outward normal to the surface, Re is the real part and the asterisk denotes the complex conjugate. Considering the entire assembly as a single "composite particle" requires to integrate the total scattered field over the whole "composite surface" and gives the scattering rate by the assembly W_s

$$W_\mathrm{s} = -\frac{1}{2}\mathrm{Re}\sum_i \oint_{S_i} [\mathbf{E}_\mathrm{s} \times \mathbf{H}_\mathrm{s}^*] \cdot \overrightarrow{\mathrm{d}s}\,. \tag{9}$$

Integrating only the wave scattered by one particular sphere over its surface and summation over all spheres would lead to neglecting the electromagnetic interaction between the particles. As can be seen from equations (7) and (8, 9) the extinction rate of the assembly can be written as

$$W_\mathrm{e} = -\frac{1}{2}\mathrm{Re}\sum_i \oint_{S_i} \{[\mathbf{E}_\mathrm{i} \times \mathbf{H}_\mathrm{s}^*] + [\mathbf{E}_\mathrm{s} \times \mathbf{H}_\mathrm{i}^*]\} \cdot \overrightarrow{\mathrm{d}s} \tag{10}$$

After some extensive mathematics we find for the extinction rate

$$\begin{aligned} W_\mathrm{e} = \frac{I_0}{|k|^2}\sum_{i,q,p}\Big[&\mathrm{Re}\left(a_{0qp}^i c_{qp}^{i*} + b_{0qp}^i d_{qp}^{i*}\right)\mathrm{Im}\left(g_1^{i,q} - g_2^{i,q}\right)\\ &-\mathrm{Im}\left(a_{0qp}^i c_{qp}^{i*} + b_{0qp}^i d_{qp}^{i*}\right)\mathrm{Re}\left(g_1^{i,q} - g_2^{i,q}\right)\\ &+2\mathrm{Re}\left(a_{0qp}^i A_{qp}^{i*} + b_{0qp}^i B_{qp}^{i*}\right)\mathrm{Im}\left(\mathrm{g}_0^{i,q}\right)\\ &-\frac{\mathrm{Im}\,(k)}{\mathrm{Re}\,(k)}\Big\{\mathrm{Re}\left(a_{0qp}^i c_{qp}^{i*} - b_{0qp}^i d_{qp}^{i*}\right)\mathrm{Re}\left(g_1^{i,q} + g_2^{i,q}\right)\\ &+\mathrm{Im}\left(a_{0qp}^i c_{qp}^{i*} - b_{0qp}^i d_{qp}^{i*}\right)\mathrm{Im}\left(g_1^{i,q} + g_2^{i,q}\right)\\ &+2\mathrm{Re}\left(a_{0qp}^i A_{qp}^{i*} - b_{0qp}^i B_{qp}^{i*}\right)\mathrm{Re}\left(\mathrm{g}_0^{i,q}\right)\Big\}\Big] \end{aligned} \tag{11}$$

and for the scattering rate

$$\begin{aligned} W_\mathrm{s} = \frac{I_0}{|k|^2}\sum_{i,q,p}\Big[&\left(\left|c_{qp}^i\right|^2 + \left|d_{qp}^i\right|^2\right)\mathrm{Im}\left(g_3^{i,q}\right)\\ &-\left(\left|A_{qp}^i\right|^2 + \left|B_{qp}^i\right|^2\right)\mathrm{Im}\left(g_0^{i,q}\right)\\ &-\mathrm{Re}\left(A_{qp}^i c_{qp}^{i*} + B_{qp}^i d_{qp}^{i*}\right)\mathrm{Im}\left(g_1^{i,q} - g_2^{i,q}\right)\\ &+\mathrm{Im}\left(A_{qp}^i c_{qp}^{i*} + B_{qp}^i d_{qp}^{i*}\right)\mathrm{Re}\left(g_1^{i,q} - g_2^{i,q}\right)\\ &-\frac{\mathrm{Im}\,(k)}{\mathrm{Re}\,(k)}\Big\{\left(\left|c_{qp}^i\right|^2 - \left|d_{qp}^i\right|^2\right)\mathrm{Re}\left(g_3^{i,q}\right)\\ &-\left(\left|A_{qp}^i\right|^2 - \left|B_{qp}^i\right|^2\right)\mathrm{Re}\left(g_0^{i,q}\right)\\ &-\mathrm{Re}\left(A_{qp}^i c_{qp}^{i*} - B_{qp}^i d_{qp}^{i*}\right)\mathrm{Re}\left(g_1^{i,q} + g_2^{i,q}\right)\\ &-\mathrm{Im}\left(A_{qp}^i c_{qp}^{i*} - B_{qp}^i d_{qp}^{i*}\right)\mathrm{Im}\left(g_1^{i,q} + g_2^{i,q}\right)\Big\}\Big]\,, \end{aligned} \tag{12}$$

where

$$g_0^{i,q} = \psi_q(\rho_i)\psi_q'^*(\rho_i)\,, \tag{13}$$

$$g_1^{i,q} = \xi_q(\rho_i)\psi_q'^*(\rho_i)\,, \tag{14}$$

$$g_2^{i,q} = \xi_q'(\rho_i)\psi_q^*(\rho_i)\,, \tag{15}$$

$$g_3^{i,q} = -\xi_q(\rho_i)\xi_q'^*(\rho_i)\,, \tag{16}$$

$$A_{qp}^i = \frac{c_{qp}^i}{\Gamma_k^i} - a_{0qp}^i\,, \tag{17}$$

$$B_{qp}^i = \frac{d_{qp}^i}{\Delta_q^i} - b_{0qp}^i\,. \tag{18}$$

Here $\psi_q(\rho) = \rho j_q(\rho)$ and $\xi_q(\rho) = \rho h_q^{(1)}(\rho)$ are Ricatti-Bessel functions [5], the prime stands for a derivative with respect to the argument, and $\rho_i = kR_i$ is the complex size parameter of the i-th particle of radius R_i in the matrix. I_0 is the intensity of the external incident field at the origin of the coordinate system, which may be chosen at the center of one of the clusters. The attenuation of this field in the matrix is described then by *the complex phases* in the expansion coefficients a_{0qp}^i and b_{0qp}^i (explicit expressions are given in equations (56) in [2], only the complex wave vector has to be introduced). Setting $A_{qp}^i = 0$ and $B_{qp}^i = 0$ in (11) and (12) leads to the expressions for the case of the *non-interacting* particles in an absorbing host, as they were given in [1]. On the other hand, for the case of a *non-absorbing* host the expressions equivalent to equation (71) in [2] for the extinction rate and equation (21) in [6] (see also equation (14b) in [7]) for the scattering rate may be

obtained from (11) and (12) by using the properties of the Ricatti-Bessel functions for real arguments.

It is convenient to define the aggregate extinction cross-section $C_{\rm e} = W_{\rm e}/I_0$. Such a normalization provides an independence of the cross-section on the aggregate position in the matrix, and the extinction rate of the cluster assembly will be then given by a simple multiplication of the extinction cross-section with the intensity of the incident light at the "aggregate origin" (actually, in the center of the coordinate system in which the extinction rate (11) is calculated).

If identical cluster aggregates are uniformly distributed over the matrix volume an extinction coefficient $\alpha_{\rm cl}$ of clusters in the matrix may be introduced similarly to the case of separate clusters (see [1])

$$\alpha_{\rm cl} = nC_{\rm e}\,, \tag{19}$$

where n is the number density of aggregates. Then, the total extinction coefficient of the system matrix+clusters is given by

$$\alpha = \alpha_0 + \alpha_{\rm cl}\,, \tag{20}$$

where α_0 is the extinction coefficient of the pure matrix. As mentioned in [1], such a description of the composite medium (matrix + clusters) *via* macroscopic extinction coefficient requires that the condition $\alpha D \ll 1$ is satisfied, where D is the aggregate characteristic size in the direction of the light propagation. If this is not the case, nevertheless, the macroscopic extinction coefficient may approximate the optical behavior of the system with a good quality.

3 Applications

3.1 Ag-cluster chain in CuPc

To show an example of accounting both the absorption in the host matrix and the coupling between the clusters, in this section we present results of the calculation of the optical extinction of a linear chain of four equal silver spherical clusters embedded in CuPc matrix. The wave vector of a plane monochromatic incident wave was assumed to be perpendicular to the chain (normal incidence, if the clusters were arranged in a layer). The diameters of the clusters were taken to be 10 nm. The distance between the clusters centers in the chain was 10.5 nm. The coupling effect was calculated up to the 2^6-polar order, *i.e.* the sums in (11) were truncated at $s = 6$. We used the optical data for silver from [8] and for CuPc from [9]. The spectra were averaged for all polarization of the incident light. The results of the calculation are shown in Figure 1 in terms of the extinction coefficient, *i.e.* $\alpha_{\rm cl}$ from (19), where the number density of cluster aggregates corresponds to the filling factor of silver in CuPc of about 20%. For the sake of comparison we also show the extinction coefficient spectrum of separate silver clusters embedded in CuPc. There is also a spectrum of the absorption coefficient of pure CuPc placed in the same Figure.

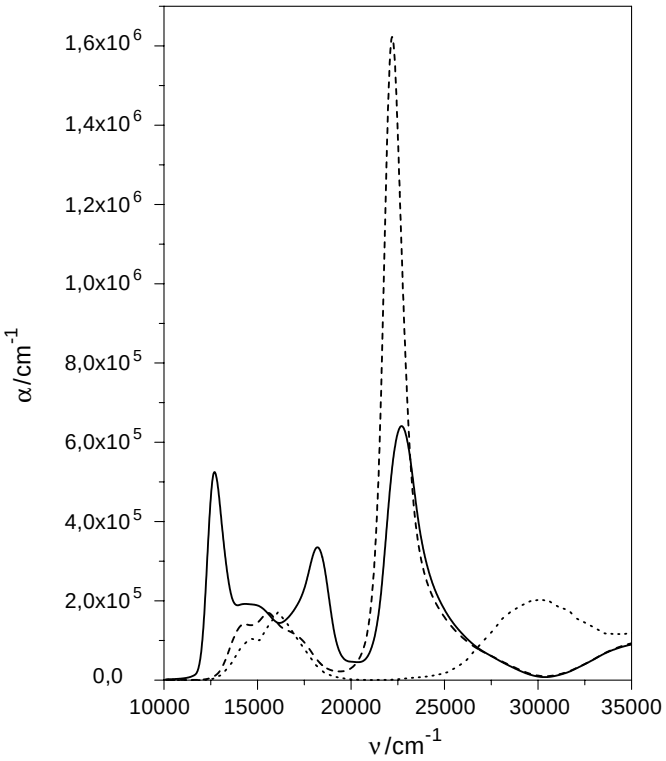

Fig. 1. Extinction coefficient of a linear chain of four spherical silver clusters embedded in CuPc averaged over all polarizations of incident light (solid curve). Extinction coefficient of separate spherical silver clusters embedded in CuPc (dashed curve). Radii of clusters are 5 nm, the inter-center distances between neighboring clusters in the chain are 10.5 nm. Number density of aggregates in the matrix corresponds to the volume filling factor of silver of about 20%. Absorption coefficient of the pure CuPc (dotted curve).

In the case of separate clusters the position of the main plasmon resonance of Ag clusters is found at 22500 $\rm cm^{-1}$, in the region of a small absorption of CuPc. The coupling between clusters gives rise to a complex structure with several resonances — at 12500 $\rm cm^{-1}$, at 18000 $\rm cm^{-1}$ and at approximately 23000 $\rm cm^{-1}$.

It must be emphasized that the observed structure is caused by both the electromagnetic coupling between clusters and the behavior of the dielectrical functions of the participating materials.

3.2 Pairs of Ag-clusters in N,N'-dimethyl-3,4,9,10-perylenedicarboximid (PTCDI)

In this section we have applied the developed method to calculate the optical response of silver clusters embedded in a PTCDI matrix. We have approximated the electromagnetic coupling (up to the octupolar order) between the clusters *via* pair interactions, where the diameter of clusters in the pair was taken to be 10 nm and the spectra were averaged over all directions of polarization of incidence wave and over distances between centers of the clus-

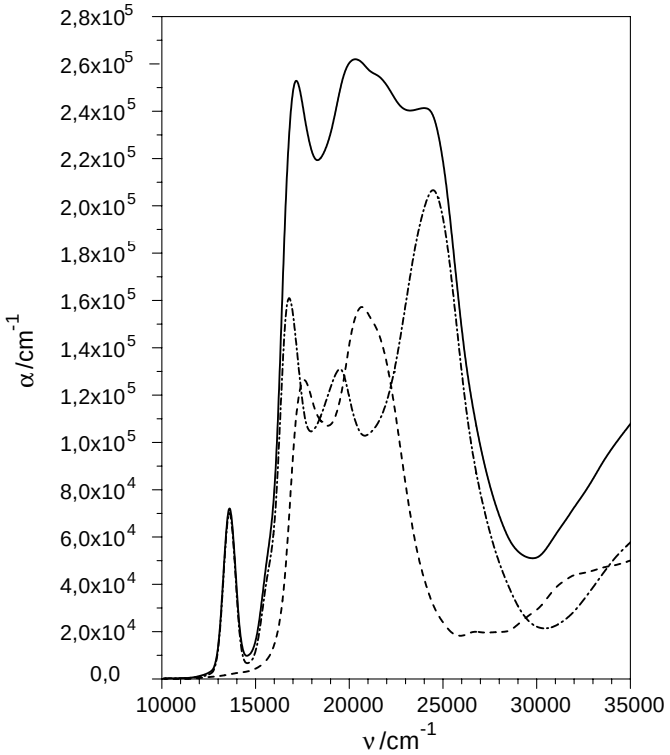

Fig. 2. Extinction coefficient of two spherical silver clusters embedded in PTCDI (dashed-dotted curve). Radii of clusters are 5 nm, the spectrum is averaged over the inter-center distances between neighboring clusters from 10 nm to 15 nm and over all directions of polarization of incident light. Number density of aggregates in the matrix corresponds to the volume filling factor of silver of about 20%. Absorption coefficient of the pure PTCDI (dashed curve). Total extinction coefficient of the composite system clusters+matrix (solid curve).

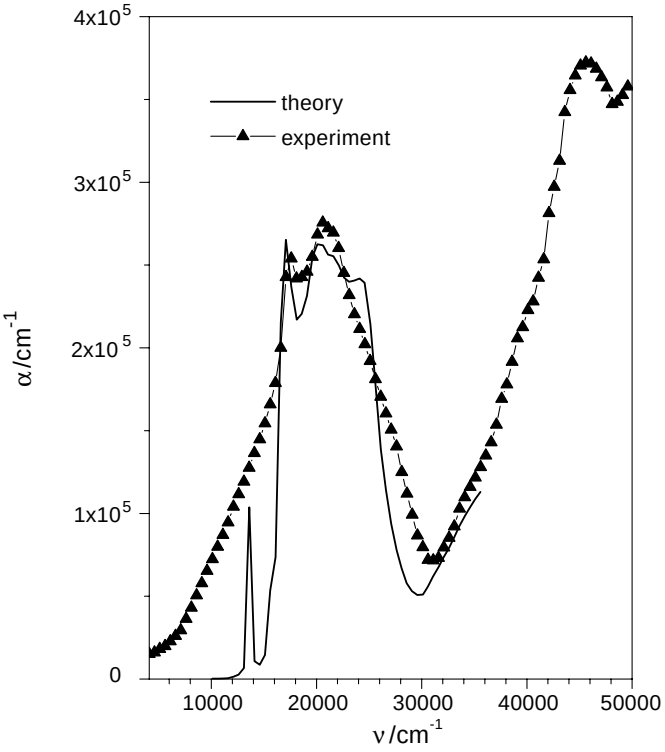

Fig. 3. Comparison of the calculated total extinction coefficient of the composite system silver clusters+PTCDI matrix (solid curve) with the experimental data (triangles).

ters from 10 nm up to 15 nm. The uniform distribution of the inter-center distances was applied for the averaging. Then, the extinction coefficient of clusters in the matrix was calculated using equation (19) and the total extinction coefficient from (20), where the number density of cluster aggregates corresponded to the volume filling factor of silver of about 20%. In Figure 2 we present these results together with the absorption coefficient of pure PTCDI. The optical data from [9] were used for PTCDI.

3.3 Comparison with experimental results

In a previous publication, we have reported on the linear optical properties (absorption behavior and Raman scattering) of noble metal clusters incorporated into CuPc- as well as PTCDI-matrices [9]. These samples had been produced by thermal evaporation, so that the clusters exhibit a random distribution of their sizes and distances. It is of course not very probable to find linear 4-cluster chains in such types of samples. Nevertheless even the idealized geometry of a 4-cluster chain leads to a very important result: the possibility of absorption in a copper phthalocyanine-silver composite at wavelength values around 800 nm (12500 cm^{-1}). This absorption is also significant in the experiment (see [9]) and cannot be explained regarding single silver spheres in the quasi-static approximation.

The simpler, but more realistic geometry of randomly oriented pairs of clusters (Section 3.2) allows a straightforward comparison with experimental data. Figure 3 shows the experimentally established extinction (absorption + scattering) coefficient of a PTCDI-silver composite with a silver filling factor of approximately 20% (taken from [9]). This curve is compared with the calculated extinction coefficient from Figure 2. The maximum values of the optical extinction coefficient are in excellent agreement. Some discrepancies between calculation and experiment occur for wave numbers around 30000 cm^{-1} and below 17000 cm^{-1}. The latter are clearly due to the simplifying assumption of uniformly sized spheres, as used in our calculation, as well as to using the uniform inter-center distances distribution for the averaging (Section 3.2). As experimental cluster sizes ranged between diameters of 3 nm to approximately 20 nm, the sharp absorption feature at 14000 cm^{-1} is expected to be superimposed with other lines originating from other cluster sizes, thus explaining the broad absorption flank as observed experimentally.

The existence of sharp silver cluster absorption lines at around 700 nm in wavelength (14000 cm^{-1}) may be an explanation for the high Surface Enhanced Raman Spectroscopy signals, which are obtained from such PTCDI-silver composites at a Raman excitation line of 647 nm (15500 cm^{-1}). Indeed, the electrodynamic SERS theory predicts a SERS efficiency which is inversely proportional to the homogeneous linewidth of the cluster absorption [9], which favors these sharp absorption features for application in SERS experiments.

4 Conclusion

The main aim of this work was to present an algorithm that allows the calculation of the full optical extinction (absorption + scattering) rate of arbitrary aggregates of spherical clusters in absorbing embedding media.

Our method is exact in the sense it is based on the rigorous theory of *Gerardy and Ausloos* and uses the exact analytical integration procedure to find the extinction rate. In terms of the presented theory under the conditions mentioned in Section 2, the full optical extinction coefficient of a matrix+cluster composite may be introduced as in the case of a macroscopically homogeneous matrix. It is calculated by adding the matrix absorption coefficient to the extinction coefficient of the cluster aggregate, which provides a relationship between experimental observables (transmission, reflection, etc.) of the sample and the aggregate extinction rate. The performed model calculations show an astonishingly good agreement with experimental results.

The advantages of our approach are, in our view, the simplicity and "transparency" of the input and output data. As input, we have the dielectric functions of the participating materials and the geometry of the aggregates (cluster sizes and distances). If a uniform distribution of cluster aggregates in the host is assumed, the volume filling factor of cluster material gives rise to the corresponding number density, and as an output, we have the absolute value of the effective extinction coefficient. In the other case (the distribution of aggregates in the host is not uniform, a layer of clusters embedded in a film, for example), if aggregates positions in the matrix are known the absolute rate of energy losses in the aggregate volume may be calculated and the total energy losses in the system may be estimated.

In contrast to simple mixing model approaches, scattering losses are incorporated in our theory. On the other hand, an integration of Poynting vector over particles surfaces may be crucial in a case of limited-size embedding media and allows to avoid an empirical postulation of core-shell particles to take into account the matrix absorption.

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References

1. A.N. Lebedev, M. Gartz, U. Kreibig, O. Stenzel, Eur. Phys. J. D **6**, 365 (1999).
2. J.M. Gerardy, M. Ausloos, Phys. Rev. B **25**, 4204 (1982).
3. M. Quinten, O. Stenzel, A. Stendal, C. von Borczyskowski, J. Opt. **28**, 245 (1997).
4. A.L. Aden, M. Kerker, J. Appl. Phys. **22**, 1242 (1951).
5. M. Abramowitz, I.A. Stegun, *Handbook of Mathematical Equations* (New York, Dover, 1972).
6. A.N. Lebedev, O. Stenzel, M. Quinten, A. Stendal, M. Roeder, M. Schreiber, D.R.T. Zahn, to be published in J. Opt. A: Pure Appl. Opt. (1999).
7. M. Quinten, U. Kreibig, Appl. Opt. **32**, 6173 (1993).
8. M. Quinten, Z. Phys. B **101**, 211 (1996).
9. O. Stenzel *et al.*, Appl. Surf. Sci. **108**, 71 (1997).