

Resonant Absorption of Electromagnetic Radiation in Matrix Disperse Systems with Metallic Inclusions

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The absorption of electromagnetic radiation in low density system of fine metallic particles embedded in dielectric matrix is considered with account of influence of dipole-dipole interaction between particles. It is shown that under account of pair interaction between particles near proper plasma surface mode of a single particle two new neighboring resonant modes appear, their excitation creating the region of continuous spectrum. In a result, the mechanism of collisionless damping occurs leading to widening and doubling of the resonance peaks in absorption. The dependence of absorption on the average distance between particles and on collision frequency is examined.

Introduction

This paper is logical development of the preceding article [1]. Disperse systems when interacting with electromagnetic radiation (EMR) reveal a number of peculiar features in absorption and scattering which are absent in analogous processes in the bulk continuous media (BCM). They appear due to the presence of supplementary factors as compared with BCM - the concentration and form of inclusions, their structure and geometry of location in a matrix and so on. We note only some peculiarities of such processes in the matrix disperse systems (MDS), i.e. in systems that consist of continuous matrix with intruded inclusions of various nature (metals, insulators etc.):

- anomalous absorption of radiation in the distant IR range (100-200 μm) in MDS with metallic inclusions[2];
- effect of the optical clearing up in MDS with two-layers dielectric inclusions [3];
- effect of enhanced Raman scattering (SERS) in colloid systems with metallic particles [4-5];
- the resonant EMR absorption in the near IR (1-10 μm) and visible range in MDS with metallic inclusions, and a number of other phenomena.

The most unexpected is the last effect for it is hard to assume how the resonant absorption becomes possible in a system whereas the EMR interacts only with conducting electrons in the inclusions. The investigation of this effect is the subject of many papers, theoretical [13-22] as well as experimental [6-12] ones, though up to now there is no complete theoretical picture that would be consistent with the experimental data.

Here, we will draw theoretical study of specific features of this effect in MDS with spherical metallic inclusions. Upon that we use the results [1] where the improved version of Maxwell-Garnett approximation (MGT) was developed to calculate the effective dielec-

tric function $\tilde{\epsilon}(\omega)$ of similar systems with account of the direct multipole interaction between particles of the system.

As before [1], we restrict ourselves with the case of the longwave approximation, i.e. the wavelength of the incident radiation $\lambda = 2\pi c/\omega$ (ω - is the radiation frequency) is large compared with all the characteristic dimensions of inclusions, as well as with the mean distance between them.

1. Resonant character of radiation absorption in MDS with metallic inclusions

The experimental researches of the optical properties of composites on the ground of MDS with metallic inclusions have drawn to establishment of some general features in processes of EMR absorption in such systems [23, 6-12].

Providing one restricts by the simplest case of Drude's frequency dependence of dielectric function of inclusions $\epsilon(\omega)$ [23] and defining the matrix permeability ϵ_0 , these features can be summed in following statements:

1) in such MDS near surface plasmon frequency of a single particle $\omega_s = \omega_p / \sqrt{3}$ (ω_p - plasma frequency of conducting electrons of inclusions) the resonant absorption of incident radiation is observed on frequency ω_s , and besides it takes place when inclusion concentration (n) is very small,

$$f = \frac{4\pi}{3} \pi r^3 n \ll 1,$$

where r is the inclusion radius;

2) the absorption peak width is proportional to decaying frequency (γ) of conducting electrons in inclusions, and the height - f/γ ;

3) upon the increase of the filling power f , the peak widening and its shift to the longwave region are observed;

4) sometimes the second peak appears which shifts to the shortwave side on the increase of f [11].

The simple use of MGT for explanation of these regularities shows that in such systems the resonance really takes place on a frequency [7]

$$\omega_s = \omega_p \sqrt{\frac{1-f}{3}}, \quad (1)$$

and the imaginary part of the effective dielectric function of the system is $\text{Im} \tilde{\epsilon}(\omega) \approx \frac{f}{\omega_s \gamma}$. Yet, the value

of the longwave shift and the peak form predicted by this theory badly fit to experiment [13-21]. Besides, the various modifications of this approach with account for nonspherical inclusion shape that were made for explanation of two absorptive peaks do not hold the criticisms [21]. Indeed, in a case of ellipsoidal inclusions with depolarization factors L_1 and L_2 ($2L_1 + L_2 = 1$) two peaks of the resonant absorption are observed [21] on frequencies

$$\Omega_{s_1, s_2}^2 = \frac{\omega_p^2}{2} \left[\left(1 - L_1 - \frac{f}{3} \right) \pm \sqrt{\left(1 - L_1 - \frac{f}{3} \right)^2 - 4(1-f)L_1(1-2L_1)} \right], \quad (2)$$

but they both are shifting to the longwave side upon the increasing of f and the double spectrum character do not quite agree with experiment. Note, the depolarization factors L_1 and L_2 in a case of particles in a form of rotational ellipsoid with halfaxis a and b are functions only of these quantities a and b [23]. The better agreement between theory and experiment was achieved by using different improved variants of approximations: theory of multiple scattering [18-19], modified variant of the mean field with using coherent potential approach [13-14], renormed MGT approach with a diagram technique [15-17] though physical cause of width asymmetry, the presence of two peaks etc. did not find an explanation in these theories. Nevertheless, from these theories it became clear that upon the increase of f the effects of direct dipole-dipole and multipole interaction between polarized inclusions of system turn out to be essential, as well as statistical properties of particles distribution in a volume and over their dimensions. Depending on proximity to the percolation threshold, the picture becomes complicated significantly and calls for considerations about fractal properties of structures that are generated [22].

In MGT approach the resonant absorption EMR on frequencies ω_s (formula (1)) in systems under consideration is a consequence of the existence of surface plasmons in separate particles, their spectrum may be found from a condition [23]

$$\epsilon'(\omega_s) + \epsilon_0 = 0. \quad (3)$$

where $\epsilon'(\omega)$ is a real part of particle's dielectrical function (DF), and ϵ_0 is the matrix DF. When imaginary part of $\epsilon(\omega)$ is small, this condition means that the particle's polarizability on frequencies ω_s [23] increases anomalously.

In paper [1] we obtained the exact solution of a problem of behaviour of two spherical particles in electrostatic field. From this solution it follows that in spectrum of surface plasmons in this case two frequencies appear, which are caused by poles in longitudinal and transverse polarizabilities of two particles (expression (23) in [1]):

$$\omega_{\parallel}^2 = \omega_s^2 \frac{\left[1 - 2\left(\frac{r}{R}\right)^3 \right]}{1 - 2\left(\frac{r}{R}\right)^3 \left(\frac{\epsilon_{\infty} - \epsilon_0}{\epsilon_{\infty} + 2\epsilon_0} \right)}; \quad (4)$$

$$\omega_{\perp}^2 = \omega_s^2 \frac{\left[1 + \left(\frac{r}{R}\right)^3 \right]}{1 + \left(\frac{r}{R}\right)^3 \left(\frac{\epsilon_{\infty} - \epsilon_0}{\epsilon_{\infty} + 2\epsilon_0} \right)};$$

where $\omega_s = \frac{\omega_p}{\sqrt{\epsilon_{\infty} + 2\epsilon_0}}$ is surface plasmon frequency of a single particle [23], and R - the distance between two particles centers. Here we assume that the DF of inclusions has a Drude form [23]

$$\epsilon(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\gamma)}, \quad (5)$$

where the decaying frequency $\gamma = \gamma_0 + \gamma_1$, γ_0 is decaying frequency in a bulk metal, and γ_1 is caused by electron collisions with the particle surface, and has an order of magnitude r/v_F (v_F is electron Fermi velocity).

2. EMR absorption in matrix disperse systems with spherical inclusions with account of pair multipole interaction

Here, in order to relate the theoretical and experimental results which are connected with plasmon spectra of radiation absorption in MDS with metallic

inclusions we consider the frequency dependence of imaginary part of effective dielectric function $\tilde{\epsilon}(\omega)$, also consider the surface plasmon spectrum and analyze the features of radiation absorptive spectra in such systems, using results of paper [1].

The effective dielectric function of such MDS in an electrostatic approximation is found from expression (27) of paper [1]

$$\frac{\tilde{\epsilon} + 2\epsilon_0}{\tilde{\epsilon} - \epsilon_0} = \frac{1 - \frac{2}{3}f \left(\frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0} \right) \ln \frac{3\epsilon + 5\epsilon_0}{2\epsilon + 6\epsilon_0}}{f \frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0}}, \quad (6)$$

where ϵ and ϵ_0 are dielectric permeabilities of inclusions and of a matrix, correspondingly, and f is the filling power of a matrix by metallic fraction. Restricting the following consideration by the case of $\epsilon_\infty = 1$ and $\epsilon_0 = 1$, from (5) and (6) we find

$$\tilde{\epsilon}(x) = 1 + \frac{3f}{1 - f - 3x^2 - \frac{2}{3}f \ln \frac{3 - 8x^2 - 8ixv}{2 - 8x^2 - 8ixv} - i3xv}, \quad (7)$$

where $x = \omega/\omega_p$, $v = \gamma/\omega_p$.

So far, as expression (7) is fundamental for the following analysis, we underline once more that it takes place under exact account of the pair dipole interaction in a system of metallic spheres, together with the fulfillment of relations (5) and (26) of paper [1]. From (7) with account for manifoldness of logarithmic function [24], we find

$$\tilde{\epsilon}' = 1 + \frac{fL(x, v, f)}{L^2(x, v, f) + N^2(x, v, f)}; \quad (8)$$

$$\tilde{\epsilon}'' = 1 + \frac{fN(x, v, f)}{L^2(x, v, f) + N^2(x, v, f)};$$

$$N(x, v, f) = x_0^2 - x^2 - \frac{1}{9}f \ln \frac{(3 - 8x^2)^2 + 64x^2v^2}{(2 - 8x^2)^2 + 64x^2v^2}; \quad (9)$$

$$L(x, v, f) = \frac{2\pi f}{9} \Theta \left(x^2; \frac{1}{4}; \frac{3}{8} \right) + xv + \frac{2}{9}f \operatorname{arctg} \frac{8xv}{(2 - 8x^2)(3 - 8x^2) + 64x^2v^2}, \quad (10)$$

and $\tilde{\epsilon}' = \operatorname{Re} \tilde{\epsilon}(\omega)$, $\tilde{\epsilon}'' = \operatorname{Im} \tilde{\epsilon}(\omega)$.

Here $x_0^2 = \frac{1-f}{3}$ is the resonant mode frequency (2) in MGT approximation, and

$$\Theta \left(x^2; \frac{1}{4}; \frac{3}{8} \right) = \Theta_0 \left(x^2 - \frac{1}{4} \right) - \Theta_0 \left(x^2 - \frac{3}{8} \right), \quad (11)$$

$$\Theta_0(x^2 - a) = \begin{cases} 1; & x^2 > a \\ 0; & x^2 < a \end{cases}$$

and $\operatorname{arctg}(y)$ is defined in the principal region $\left(-\frac{\pi}{2}; \frac{\pi}{2} \right)$. From view of (8) it follows the important

fact: near frequency x_0^2 , which corresponds to the resonant frequency of MGT approximation, in the frequency interval $\frac{1}{4} < x^2 < \frac{3}{8}$ the continuous spectrum region appears which brings to the function

$N(x, v, f)$ the supplementary damping of an order of $\frac{2}{9}\pi f$, which does not depend on v and x ! It means

that even when $v = 0$ for any frequency $x = \omega/\omega_p$

from interval (0,5; 0,61) $\operatorname{Im} \tilde{\epsilon}(\omega) \neq 0$. In a sense

this fact reminds the mechanism of Landau damping in a collisionless plasmas. The similar mechanism was found also by calculation of the susceptibility of fractal structure with account of the pair fluctuational dipole interaction in a paper [25], and slightly had been noted in a paper [16]. Because of importance of this problem we will carry out the detailed qualitative analysis of found expressions (8)-(11). We fix our attention on analysis of dependence of $\tilde{\epsilon}''(x)$ on

frequency and on parameters v and f , because it is, namely, the imaginary part of $\tilde{\epsilon}(x)$ that is responsible for all EMR absorptive processes in a system. The exclusiveness of a given situation lies in that for the majority of disperse systems investigated practically from this point of view [6-12] the metals used as the filling matter had values $v \approx 0,01 - 0,2$, and the magnitude of f had also been lying in the same limits $f \approx 0,01 - 0,2$. At the same time, as follows from

(8), namely when $f \approx v$ the dependence $\tilde{\epsilon}''(x)$ is the most intricate. From an appearance of $\tilde{\epsilon}''(x)$ it is seen that its magnitude increases highly for the values of $x_i (i = 1, 2, \dots)$ where

$$L(x_i, v, f) = 0. \quad (12)$$

The equation (12) defines (when $v \rightarrow 0$) the surface plasmon spectrum in a system. When neglecting the pair dipole-dipole (PDD) interaction, this spectrum is determined by a single mode x_0 equal to

$$x_0 = \sqrt{\frac{1-f}{3}} \quad (13)$$

When "switching on" the PDD interaction the surface plasmon spectrum becomes complicated significantly depending on values of parameters ν and f . Because of transcendental character of equation (12), one can find values x_i only numerically, yet some estimates of their localization and conditions of appearance may be obtained from a graphical analysis of (12) when the expression (12) is written in a form divided over parameters ν and f :

$$\frac{9}{f}(x_0^2 - x^2) = \ln \frac{(3-8x^2)^2 + 64x^2\nu^2}{(2-8x^2)^2 + 64x^2\nu^2} \quad (14)$$

For convenience of the following analysis let us fix on some level $\nu = \nu_0$ and examine the dependence $L(x)$ and $\tilde{\epsilon}''(x)$ on alteration of the filling power f . When f is very small, from (14) it follows that its solution is unique, and x_i lies in continuous spectrum and coincides practically with x_0 . Despite this, when ν and f are such that the relation

$$x_i\nu_0 \gg \frac{2\pi}{9}f \quad (15)$$

is fulfilled, the dependence $\tilde{\epsilon}''(x)$ will have a sharp peak at $x = x_i$ with a height proportional to $\frac{f}{x_i\nu_0}$.

The fulfillment of (15) means that the damping in a single particle exceeds the collective effect of damping in a system, caused by PDD interaction. Upon f increase and when its values are less some boundary value f_+ (which would be determined below) the solution x_i of (14) will be unique and will lie in continuous spectrum but it will be shifted to the smaller value side of x (the "red shift"); upon this the $\tilde{\epsilon}''(x_i)$ peak will broaden significantly due to the increase of the second damping mechanism input. Such a picture would be observed in a system until f would not reach the boundary value

$$f_+ = \frac{0,25}{1 + \frac{1}{3} \ln \left(\frac{1+16\nu^2}{16\nu^2} \right)} \quad (16)$$

When $f \geq f_+$ there appears in a system near $x \approx 1/4$ the supplementary root, and upon the following increase of f their number extends to three, where one of roots lies in a region $x_i < 1/4$, and two

others x_2 and x_3 - in a region of continuous spectra. The originated mode x_i will be narrow as its half-width is proportional to the damping ν , and the height is $\frac{f}{x_i\nu}$. Upon the following increase of f the modes inside continuous spectra disappear, there leaves only a mode x_i which acquires the longwave shift (the "red shift"). Under the still prolonging increase f , when $f > f_-$, where

$$f_- = \frac{0,125}{\frac{1}{3} \ln \left(\frac{1+24\nu^2}{24\nu^2} \right) - 1} \quad (17)$$

two new modes, besides x_i , are born in a system near $x = 3/8 - x_2$ and x_3 , where x_2 lies in a continuous spectrum, and x_3 - in a region $x > 3/8$, i.e. the mode x_3 is the resonant one. In distinction from x_i , the mode x_3 has a shift to the shortwave side ("blue shift"). Note, that the mode x_2 lying in continuous spectrum experiences the longwave shift. But, as follows from analysis of expression (12) modes x_2 and x_3 are located near value $x = 3/8$ and decay more strongly than the mode x_i . The evolution picture of $\text{Im} \tilde{\epsilon}(x)$ dependence of changing on parameter f from 0,001 to 0,3 ($\nu=0,01$) is shown in Fig. 1 and Fig. 2 justify completely the analysis drawn over solution of equation (12). Indeed, as follows from (16) and (17), by $\nu=0,01$ the mode x_i appears when $f \approx 0,08$, and the mode x_3 when $f \approx 0,125$. That is correlated completely with results shown in Fig. 1 and Fig. 2. The evolution of $\text{Im} \tilde{\epsilon}(x)$ dependence under fixed $f \approx 0,09$ and alteration ν from 0,01 to 0,1 is shown in Fig. 3 and Fig. 4. From (16) in this case follows that resonant mode x_i appears when $\nu \leq 0,02$ that agrees well with results of Fig. 3. When $\nu > 0,02$, in the spectrum there are present only modes inside the continuous spectrum which are giving due to the damping mechanism specific in this interval the asymmetric bell-like curves of dependences of $\text{Im} \tilde{\epsilon}(x)$. In Fig. 5 the frequency dependences of the absorption coefficient are shown:

$$\alpha = \frac{4\pi c}{\omega} \text{Im} \sqrt{\tilde{\epsilon}(\omega)} \quad (18)$$

when $\nu=0$ and f changing from 0,01 to 0,1.

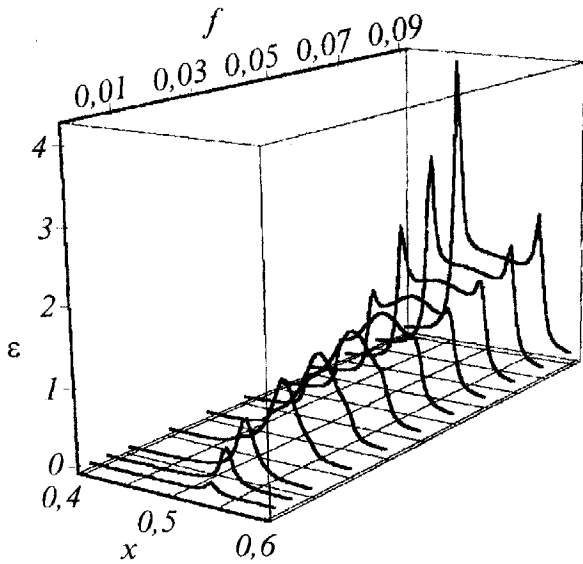


Fig. 1. Dependence $\text{Im } \tilde{\epsilon}$ on $x = \omega/\omega_p$ under damping $\nu = \gamma/\omega_p = 0,01$ for different filling powers $f=0,01 \div 0,09$

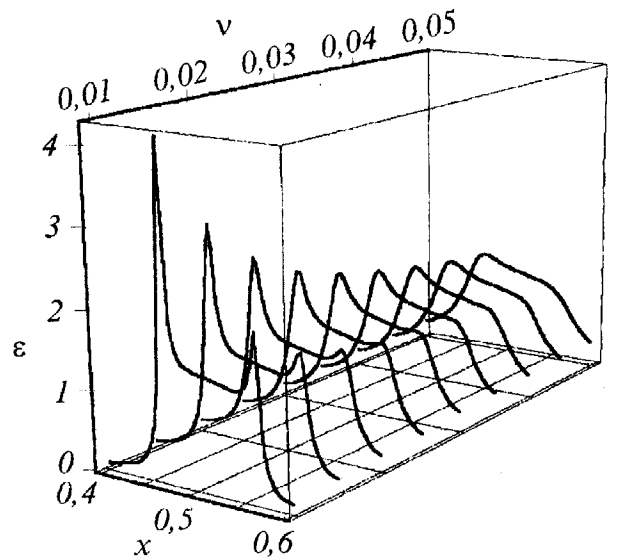


Fig. 3. Dependence $\text{Im } \tilde{\epsilon}$ on $x = \omega/\omega_p$ under filling power $f=0,09$ for different damping values $\nu = \gamma/\omega_p = 0,01 \div 0,05$

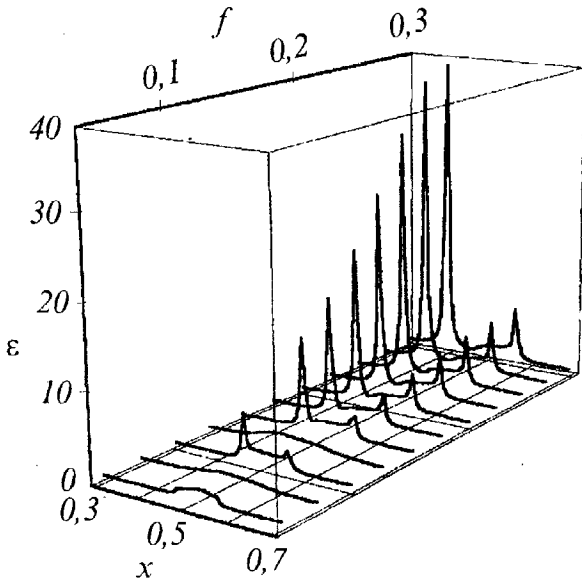


Fig. 2. Dependence $\text{Im } \tilde{\epsilon}$ on $x = \omega/\omega_p$ under damping $\nu = \gamma/\omega_p = 0,01$ for different filling powers $f=0,09 \div 0,3$

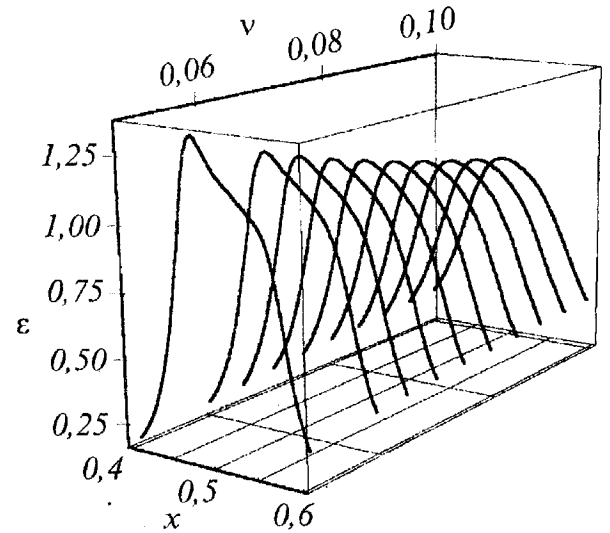


Fig. 4. Dependence $\text{Im } \tilde{\epsilon}$ on $x = \omega/\omega_p$ under filling power $f=0,09$ for different damping values $\nu = \gamma/\omega_p = 0,05 \div 0,1$

Note, that the origin of continuous spectrum region is observed also in general case, when the dielectric permeability $\epsilon_0 \neq 1$, and $\epsilon(\omega)$ dependence has a form (5). In this case the continuous spectrum region is in interval

$$\frac{3}{3\epsilon_0 + 5\epsilon_\infty} < x^2 < \frac{1}{\epsilon_0 + 3\epsilon_\infty} \quad (19)$$

where spectrum width is equal to

$$\Delta = \frac{8\epsilon^-}{(3\epsilon_0 + 5\epsilon_\infty)(\epsilon_0 + 3\epsilon_\infty)} \quad (20)$$

From (19) it follows that Δ is maximal when $\epsilon_0 = \epsilon_\infty = 1$.

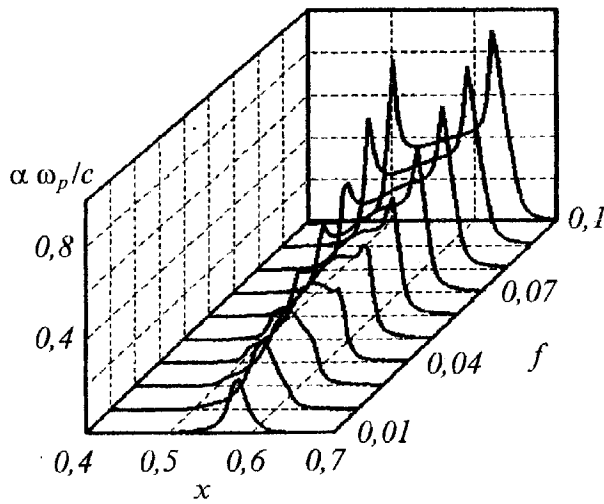


Fig. 5. Dependence of absorptivity $\alpha\omega_p/c$ on $x = \omega/\omega_p$ under damping $\nu = \gamma/\omega_p = 0,01$ for different filling powers $f=0,01 \div 0,1$

3. Discussion of results

The calculations that were drawn show that the account of pair DD interaction between the metallic inclusions in a system turns to the appearance of the continuous spectrum band in the region of surface plasmas mode of an separate inclusion $(\omega_p/\sqrt{3})$.

This spectrum region appearance is explained by that in the case of the account of the pair interaction between inclusions any two particles belonging to the system absorb the interaction on frequencies ω_{\parallel} and

ω_{\perp} , which are defined by relations (4). These frequencies values depend on distance between particles centers R . So, when $R=\infty$, in a case $\epsilon_0 = \epsilon_{\infty} = 1$,

$\omega_{\parallel} = \omega_{\perp} = \omega_p/\sqrt{3}$. Under the minimal approach

of particles $R=2r$, $\omega_{\parallel}^* = \frac{\omega_p}{2}$, $\omega_{\perp}^* = \sqrt{\frac{3}{8}}\omega_p$, i.e.

these frequencies just determine the boundaries of the continuous spectrum. As R is arbitrary, it becomes possible in a system the presence of a wide frequency set. It turns to the possibility of quick relaxation of separate particle polarizability in the continuous spectrum region which does not depend on radiation absorption character in a single particle, i.e. it appears the supplementary damping mechanism indicated before. This consequence has, by semblance, the general nature. The origin of modes outside the continuous spectrum region is connected only with dipole-dipole approach which takes place in the case when the average distances between particles $\bar{R} > 3r$ [26]. It turns to that the given approximation is true only

when $f < 0,15$. When $R < 3r$, the highest multipole interactions (quadrupole, octupole and so on) become significant [27]. The estimates drawn have shown that the account of these interactions turns to the significant broadening of the continuous spectrum region, so the separate modes outside the continuous spectrum boundaries in dipole-dipole approach get into the continuous spectrum region, its boundaries are found from the approximation with account of quadrupole interaction and so on. In that sense one should pay attention to the paper [20] where the radiation absorption band contour was analyzed in ensemble of small metallic particles. For calculation of the absorption coefficient in a paper [20] the effective dielectric permeability was used, found by formula

$$\tilde{\epsilon} = \epsilon_0 \frac{1 + 2f\theta}{1 - f\theta}, \quad (21)$$

$$\theta = \left(\frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0} \right) \left[1 + G \left(\frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0} \right)^2 \right]. \quad (22)$$

where G is the phenomenological parameter of the theory, which depends on the sphere radius, the filling factor and on the relative S -particle ($S = 1, 2, 3, \dots$) correlative functions. Further, in [20] the alteration of absorption bands is studied under changing of a parameter G from 0 to 10. In spite of the fact that the significant change of the absorption band width by $G > 1$ was observed, all the theory remains fully phenomenological because the nature of such widening is left unclear in [20].

Note, that the formula analogous to (21) can be easily obtained from a relation (7) by expansion of a logarithm into series over parameter $A = \frac{\epsilon - \epsilon_0}{\epsilon + 2\epsilon_0}$,

though it would turn out that the introduced quantity G should depend also on $\epsilon(\omega)$ and ϵ_0 , i.e. on radiation frequency.

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Резонансное поглощение электромагнитного излучения в матричных дисперсных системах с металлическими включениями

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Исследовано поглощение электромагнитного излучения в системе малых металлических частиц, растворенных в диэлектрической матрице, с учетом диполь-дипольного взаимодействия между частицами. Показано, что учет парного взаимодействия между частицами приводит к появлению вблизи частоты собственной плазменной поверхностной моды отдельной частицы двух новых резонансных мод, создающих область непрерывного спектра. В результате становится возможным механизм бесстолкновительного затухания, который приводит к удвоению и уширению резонансных пиков поглощения. Изучена зависимость формы поглощения от среднего расстояния между частицами и элетронной частоты столкновений.

Резонансне поглинання електромагнітного випромінювання в матричних дисперсних системах з металевими включеннями

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Досліджено поглинання електромагнітного випромінювання у системі малих металічних частинок, розчинених у діелектричній матриці, з урахуванням диполь-дипольної взаємодії між частинками. Показано, що урахування парної взаємодії між частинками призводить до появи поблизу власної плазменної поверхневої моди окремої частинки двох нових резонансних мод, які створюють область неперервного спектру. Внаслідок цього стає можливим механізм беззіткненого згасання, який призводить до подвоєння та розширення резонансних піків поглинання. Вивчено залежність форми поглинання від середньої відстані між частинками та електронної частоти зіткнень.