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Linear and Nonlinear Local Resonances in Thin Rough Films

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ABSTRACT

In a thin dielectric, semiconductor or metal film in the regions or indentation there exist local "Plasmon" resonances of small radius ' $t_n \ll L$, where L is the "horizontal" size of the roughness. Main characteristics of these resonances (frequencies and widths) are discussed as dependent on the roughness and on the degree of the film indentation when the dependence of the dielectric constant of the film on the electric field strength ($\varepsilon = \varepsilon_0(w) + \alpha E^2$) in the region of the "Plasmon" resonance (i.e. at $|\varepsilon_0(w)| \ll I$) is taken into account, it leads to the appearance of new ("non-linear") local resonances, The conditions of the appearance of such resonances, specially, the region of their localization, differ from analogous conditions for linear resonances.

INTRODUCTION

In this paper, some peculiarities of the spectrum of local resonance in thin rough films are discussed within the framework of macroscopic electrodynamics. We assume that the film thickness d is large compared to the characteristic microscopic lengths (the lattice constant the length of the Debye screening, the electron free path etc.) at which the value of the local dielectric constant can be established. Thus, the film may be considered to be microscopic at the same time the film thickness is assumed small compared to the field wavelength in the film. Therefore, inside the film we may neglect variation of the field across its thickness and thus the form of the basic equations for the field becomes considerably simpler that of analogous equations for bulk samples.

Next assumption which also simplifies the theory refers to the inclusion of retardation, We shall not take it into account, assuming that the radii of the considered local states in the film are wavelength of radiation ($\lambda_o = \frac{2\pi C}{\omega_o}$, where ω_o is the frequency of a local resonance). [1]

Usually localization radii of such states are small compared to the approximation of the order of the "horizontal" size of roughness L and the above said means that $L \ll \lambda_0$. If the film covers a smooth surface Z = 0 only upper surface of the film may be considered rough. In this case the film thickness at different points x, y is, generally speaking, different and it may be characterized by the function $Z = \tau(x, y)$. We assumed that the medium over the film has the dielectric constant of $\varepsilon = 1$ (vacuum), that the local dielectric constant of the film is $\varepsilon_1(\omega)$ and that the dielectric constant of the substance is $\varepsilon_2(\omega)$. Assuming roughness to be sufficiently smooth, we also assume that $|\nabla \tau| \ll 1$.

Upon neglecting retardation (i.e. within the limit $C \to \infty$, C is the velocity of light) the Maxwell equations for the electric field \overrightarrow{E} (r, t) are known to have the form:

 $div \vec{D} = 0$

 $r_{a}t\vec{E}=0.$

where \overrightarrow{D} is the electric displacement vector. Therefore, $\overrightarrow{E} = -\nabla_{\Psi} (r, t)\Delta_{\Psi} = 0$ and for the fields with frequency ω , $(\overrightarrow{E}(r, t) = \overrightarrow{E}(r)|t|e^{iwt}$, etc.) $\overrightarrow{D}(r) = \in (\omega)\overrightarrow{E}(\overrightarrow{r})$.

Thus, within the electrostatic limit under discussion, we must solve the Laplace equation $\Delta \Psi(\vec{r}) = 0$ of the three layered system being considered. The main difficulties are caused by the necessity to provide the known boundary conditions (Continuity of the field E_t and D_n ; Indices n and t denote the normal and the tangential components of the field) to be fulfilled, across the rough surface in particular.

THEORETICAL CONSIDERATION AND CALCULATIONS

The expressions for the potential $\Psi(\mathbf{r})$ in vacuum, in the film and in the substrate in the following form:

$$\Psi(\vec{\hat{\rho}}, \vec{Z}) = \sum_{K} \Psi(\vec{K}) e^{ikp \cdot \vec{K}|Z}, \vec{Z} > \tau |\vec{\hat{\rho}}|$$

$$\Psi_{1}(\vec{\hat{\rho}}, \vec{Z}) = \sum_{K} e^{ikp} \{ \Psi_{1}(\vec{K}) e^{\vec{K}|Z} + \Psi_{1}(\vec{K}) e^{-\vec{K}|Z} \}, \tau |\vec{\hat{\rho}}| > \vec{Z} > 0 \Rightarrow$$

$$\Psi_{2}(\vec{\hat{\rho}}, \vec{Z}) = \sum_{K} e^{ikp} \Psi_{2}(\vec{K}) e^{|K|Z^{-1}} \vec{Z} < 0 \qquad (1)$$

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where $\overrightarrow{p}(x, y)$ and $\overrightarrow{k} = (k_x, k_y)$ are two dimensional position and wave vectors, respectively. Since the function $e^{i\overrightarrow{k}\overrightarrow{p} + |\overrightarrow{k}|Z}$ at any real k satisfies the Laplace equation the expressions written out in equation (1) also satisfy this equation. The function $\Psi(\overrightarrow{k})$, $\Psi_1(\overrightarrow{k})$ and $\Psi_2(\overrightarrow{k})$ in these expression must be determined afterward in such a way that the above mentioned boundary conditions for the fields be fulfilled (at $|\overrightarrow{Z}| \rightarrow \infty$ the potentials Ψ and Ψ_2 tend to zero thus providing localization of the field near the film). In order to obtain the equation for the functions $\Psi, \Psi_2, \overline{\Psi}_1, \overline{\Psi}_2$ we write out in the explicit from the expression for the components of the electric field strength: in vacuum (i.e. at $\overrightarrow{Z} > \tau(\overrightarrow{p})$)

$$E_{xy}^{v}(\rho, \boldsymbol{Z}) = -i\sum_{K} K_{xy} \Psi(\vec{k}) e^{iK\rho - |K|\boldsymbol{Z}},$$

$$E_{z}^{v}(\vec{\rho}, \boldsymbol{Z}) = \sum_{K} |K| \Psi(\vec{k}) e^{i\vec{k}\rho - |\vec{k}|\boldsymbol{Z}},$$
(2a)

In the film (i.e. at $\tau(\vec{p}) > \mathbb{Z} > 0$)

$$E_{xy}^{f}(\vec{\rho}, \mathbf{Z}) = -i\sum_{K} K_{xy} e^{i\vec{k}\rho} \{ \Psi_{1}(\vec{k}) e^{|K|\mathbf{Z}} + \Psi_{1}(K) e^{|K|\mathbf{Z}} \}$$

$$E_{z}^{f}(\vec{\rho}, \mathbf{Z}) = -i\sum_{K} (\vec{k})_{xy} e^{iK\rho} \{ \Psi_{1}(\vec{k}) e^{i\vec{k}|\mathbf{Z}} + \Psi_{1}(\vec{k}) e^{i\vec{k}|\mathbf{Z}} \}$$

$$(2b)$$

In the substrate (i.e. at Z < 0)

$$E_{xy}^{s}(\vec{\rho}, \mathbf{Z}) = -i\sum_{K} K_{xy} \Psi_{2}(\vec{k}) e^{ikp + |\vec{k}|\mathbf{Z}}$$

$$E_{z}^{x}(\vec{\rho}, \mathbf{Z}) = -i\sum_{K} |\vec{k}| \Psi_{2}(\vec{k}) e^{i\vec{k}p + |\vec{k}|\mathbf{Z}}$$
(2c)

First we consider the film substrate interface ($\mathbf{Z} = 0$). From the continuity condition for E_{xy} i.e. from the condition $E_{xy}(\vec{\rho}, 0) = E_{xy}^{f}(\vec{\rho}, 0)$ we find

$$\Psi_2(\vec{k}) = \Psi_1(\vec{k}) + \Psi(\vec{k}) \tag{3}$$

If now we require continuity of the normal component of the electric displacement vector D_n , then from the condition $\in_2 E_z(\vec{p}, 0) = \in_1 E_2^f(\vec{p}, 0)$

We obtain

$$\in_1 \left[\Psi_1(\vec{k}) - \Psi_1(\vec{k}) = \in_2 \Psi_2(\vec{k}) \right] \tag{4}$$

To obtain analogous relationship for the film-vacuum interface one should take into account that on this interface the unit vector of the normal to the surface $\overrightarrow{\eta}(x, y)$ is not directed along the axis z and accordingly the tangential unit vector \in_1, \in_2 are no more parallel to the plane (x, y). In this case unit vector of the normal directed into vacuum is determined by the known relationship

$$\overrightarrow{\eta} = \left(\frac{\partial \tau}{\partial x}, \frac{\partial \tau}{\partial y}, 1\right] [1 + |\nabla \tau|^2]^{-1/2}$$





Figure 1: Function of the film profile so that to the first order of small gradients $|\nabla \tau| < < 1$

$$\vec{\eta} = \left(\frac{\partial \tau}{\partial x}, -\frac{\partial \tau}{\partial y}, 1\right)$$
(5)

In the same approximation two tangential unit vectors are written out below:

$$t_{1} = \begin{pmatrix} 1; 0; & -\partial \tau \\ \partial x \end{pmatrix}$$

$$t_{2} = \begin{pmatrix} 0; 1; & -\partial \tau \\ \partial y \end{pmatrix}$$
(6a)
(6b)

It is clear that the direction of the vector t_1 is close to that of the axis x, whereas the direction of t_2 is close to that of the y axis. According to equation (5) the normal electric displacement component at $D_n = (Dn)$ is determined by the relationship.

$$D_{n} = D_{z} = D_{x} \frac{-\partial \tau}{\partial x} - D_{y} \frac{-\partial \tau}{\partial y} = D_{z} - \frac{D\tau}{\partial \overrightarrow{\rho}} D\overrightarrow{\rho}$$
(7)

In this relationship two components of electric displacement vector are taken on the film surface. Since film is assumed to be thin, the approximation

$$\begin{split} & \mathbf{D}_{\mathbf{z}}(\overrightarrow{\boldsymbol{\rho}}, \ \vec{\tau}) = \mathbf{D}_{\mathbf{z}}(\overrightarrow{\boldsymbol{\rho}}, \ 0) + \ \vec{\tau}(\overrightarrow{\mathbf{P}}) \frac{\partial \mathbf{D}_{\mathbf{z}}}{\partial \mathbf{Z}} \bigg|_{\mathbf{z} = 0} \\ & \mathbf{D}_{\mathbf{x}}(\overrightarrow{\boldsymbol{\rho}}, \ \vec{\tau}) = \mathbf{D}_{\mathbf{x}}(\overrightarrow{\boldsymbol{\rho}}, \ 0), \ \mathbf{D}_{\mathbf{y}}(\overrightarrow{\boldsymbol{\rho}}, \ \vec{\tau}) = \mathbf{D}_{\mathbf{y}}(\boldsymbol{\rho}, \ 0), \end{split}$$

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must be used in equation (7) to an accuracy as above. If now we recall the equation div D = 0 and thus,

take into account that

$$\frac{\partial D_{z}}{\partial z} = -\frac{\partial D_{x}}{\partial x} - \frac{\partial D_{y}}{\partial y} = -\frac{\partial D_{z}}{\partial z}$$

Then equation (7) may also be written in the form

$$D_{\overline{\uparrow}}(\overline{\rho},\rho) = D_{z}(\overline{\rho},0) - \frac{\partial}{\partial \overline{\rho}} \{\tau(\overline{\rho}) D_{\rho}(\overline{\rho},0)\}$$
(7a)

In this relationship the normal electric displacement component on the outer surface of the film (i.e, at Z =

 $\tau(\rho)$) is expressed through its value at $\mathbf{Z} = 0$

In vacuum, according to equation (2a)

$$D_{\widehat{\eta}}(\widehat{\rho}, t) = \sum_{k} |\widehat{K}| \Psi(k) \ell^{ikp} + \frac{i\partial}{\partial x} \left\{ \tau \sum_{k} k_{x} \Psi(\widehat{k}) \ell^{ikp} \right\} + \frac{i\partial}{\partial y} \left\{ \tau \sum_{k} k_{x} \Psi(\widehat{k}) \ell^{ikp} \right\}$$
$$= \sum_{k} \Psi(\widehat{k}) \ell^{ikp} \left\{ |\widehat{k}| + i \left[k_{x} \frac{\partial \tau}{\partial x} + k_{y} \frac{\partial \tau}{\partial y} \right] - \tau k^{2} \right\} = \sum_{k} \Psi(\widehat{k}) \ell^{ikp} |\widehat{k}|$$

Doing so we have neglected the terms of the order of $|\vec{k}| \tau$ and $|\nabla \tau|$. since $|\nabla \tau| \ll 1$ and also, according to the assumption, the main contribution to D_{η} comes from \vec{k} for which kd $\ll 1$, using equation (7a) and (2b) we find analogously that the normal electric displacement at component inside the film

$$\begin{split} D_{\eta}(\overrightarrow{p}, \mathbf{z} = \mathbf{t}) &= - \in {}_{1}(\omega) \left\{ \sum_{l} (\overrightarrow{k}) \ell^{ikp} \left(\Psi_{l} - \overrightarrow{\Psi}_{l} \right) + i \frac{\partial}{\partial \mathbf{x}} \left[\mathbf{t} \sum_{k} \ell^{ikp} k (\Psi_{l} + \overrightarrow{\Psi}_{l}) \right] \\ &+ \frac{\partial}{\partial \mathbf{y}} \left[\mathbf{t} \sum_{k} \ell^{ikp} k_{y} (\Psi_{l} + \overrightarrow{\Psi}_{l}) \right] \right\} \end{split}$$

or, if we take into account (3) and (4)

$$D^{f}_{\eta}(\vec{p}, \tau) = - \in \sum_{k} (\vec{k}) \Psi_{2}(\vec{k}) \ell^{ikp} + \in \operatorname{idiv} \left\{ \tau(\vec{p}) \sum_{k} \vec{k} \ell^{ikp} \Psi_{2}(\vec{k}) \right\}$$

No equating $D^{v}_{\eta}(\vec{p}, t)$ with $D^{f}_{\eta}(\vec{p}, t)$ as expressed above, we obtain the equation

$$\sum_{k} \Psi(\vec{k}) \ell^{ikp}(\vec{k}) = - \bigoplus_{k} \sum_{k} |\Psi_2(\vec{k})| \ell^{ikp} + i \in I \text{ idiv } \left\{ \tau(\vec{\rho}) \sum_{k} k \ell^{ikp} \Psi_2(\vec{k}) \right\}$$
(8)

To obtain the last of the relationships which we need, we shall use the continuity of tangential field components on the film surface, since, according to equation (6a)

$$\mathrm{E}\,\tau_{\mathrm{I}}\left(\vec{\rho},\ \tau\right) = \vec{\tau}_{\mathrm{I}}\vec{\mathrm{E}}(\vec{\rho},\ t) = \mathrm{E}_{\mathrm{x}}\left(\vec{\rho},\ t\right) + \frac{\partial\tau}{\partial x}\mathrm{E}_{\mathrm{z}}\left(\vec{\rho},\ t\right)$$

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We obtain analogously with the above procedure and with the required accuracy.

$$Et_{1}(\vec{\rho}, \tau) = E_{x}(\vec{\rho}, 0) + \tau \frac{\partial \boldsymbol{E}_{x}}{\partial \boldsymbol{z}} \Big|_{\boldsymbol{z}=0} + \frac{\partial \tau}{\partial \boldsymbol{z}} E_{z}(\vec{\rho}, 0)$$

From $\omega t \vec{E} = 0$ it follows that

$$\frac{\partial E_z}{\partial x} = \frac{\partial E_x}{\partial z}$$

and thus

$$Et_{1}(\vec{\rho}, \tau) = E_{x}(\vec{\rho}, 0) + \tau \frac{\partial}{\partial x} \{\tau(\vec{\rho})E_{z}(\vec{\rho}, 0)\}$$
(9a)

In the same way we find that

$$Et_{2}(\vec{\rho}, \vec{\tau}) = E_{y}(\vec{\rho}, 0) + \frac{\partial}{\partial \chi} \{ \pi(\vec{\rho}) E_{z}(\vec{\rho}, 0) \}$$
(9b)

In vacuum, according to equation (9) and (2a)

$$E^{v}t_{1}(\vec{p}, \tau) = -i\sum_{k} k_{x}\Psi(\vec{k})\ell^{ikp} + \frac{\partial}{\partial x} \{\tau(\vec{p})\sum_{k}(\vec{k})\Psi(k)\ell^{ikp}\}$$
$$E^{v}_{t2}(\vec{p}, t) = -i\sum_{k} k_{x}\Psi(\vec{k})\ell^{ikp} + \frac{\partial}{\partial x} \{t(\vec{p})\sum_{k}(\vec{k})\Psi(\vec{k})\ell^{ikp}\}$$

and again we neglect for the reasons given above small values of the order of $\tau |\mathbf{k}|$ and $|\mathbf{v}\tau|$ compared to unity and obtain finally

$$E_{t1}^{v}(\vec{\rho}, \vec{\tau}) = -\sum_{k} i\Psi(\vec{k})\ell^{ikp}\vec{k}_{x}$$

$$E_{t2}^{v}(\vec{\rho}, \vec{\tau}) = -\sum_{k} i\Psi(\vec{k})\ell^{ikp}\vec{k}_{y}$$
(10)

Using now (9a, b) and also (2b), (3) and (4) we find that inside the film

$$E_{t1}^{f}(\vec{p}, t) = -i\sum_{k} k_{x} \Psi_{2}(\vec{k}) \ell^{ikp} - \frac{\partial}{\partial x} \left\{ \vec{p} \right\} \frac{\epsilon_{2}}{\epsilon_{1}} \sum_{k} \vec{k} |\Psi_{2}(\vec{k})\Psi_{2}(\vec{k}) \ell^{ikp} \right\}$$

and analogously for E_{t2}^{f} . Thus, the last of the relationships we require are

$$\begin{split} \sum_{k} \Psi(\vec{k}) \ell^{ikp}(-ik_{x}) &= -i\sum_{k} k_{x} \Psi_{2}(\vec{k}) \ell^{ikp} - \frac{\partial}{\partial x} \left\{ \frac{\epsilon_{2}}{\epsilon_{1}} \tau(\vec{p}) \sum_{k} |\vec{k}|(\vec{k}) \ell^{ikp} \right\} \\ \sum_{k} \Psi(\vec{k}) \ell^{ikp} k &= \sum_{k} k \Psi_{2}(\vec{k}) \ell^{ikp} - \vec{\nabla} \left\{ \frac{\epsilon_{2}}{\epsilon_{1}} \tau(\rho) \sum_{k} |\vec{k}| \Psi_{2}(\vec{k}) \ell^{ikp} \right\} \end{split}$$
(11)

We shall be interested below in the region of frequencies $\omega \Omega \omega_0$, where ω_0 is the frequency of longitudinal field vibration in the film material $\epsilon_1(\omega) = 0$. For the stated frequency region the second term

in the right-hand side of equation (8) may be considered to be small. If it is omitted, it follows directly from equation (8) that approximately

$$\Psi_2(\vec{k}) = - \frac{1}{\epsilon_2} \Psi(k)$$

Using this relationship together with equation (11) we find that the function $\Psi(k)$ which determines the field in vacuum over the rough surface of the film (see in figure 1) satisfies the following integral equation:

$$-i\sum_{k} \widehat{k}\Psi_{2}(k)\ell^{ikp}\left[1+\frac{1}{\epsilon}\right] = \frac{1}{\epsilon}\int_{1} \widehat{\nabla}\left\{\tau(\rho)\sum_{k} \widehat{k}\Psi(k)\ell^{ikp}\right\}$$

This equation may also be rewritten as $\vec{\nabla} E(\vec{p}) = \sigma$

where
$$F(\vec{p}) = \sum_{k} \Psi(\vec{k}) \ell^{ikp} \left[1 + \frac{1}{\epsilon} \right] + \frac{\tau(\vec{p})}{\epsilon_{1}} \sum_{k} |\vec{k}| \Psi(\vec{k}) \ell^{\vec{k}p}$$

Thus, $F(\vec{p}) = \text{constant}$. This constant is determined by the choice of the potential value at infinity and therefore it may be assumed to be zero. Consequently, the equation for the function $\Psi(k)$ may be written \Rightarrow in its final form as

$$\left[1 + \frac{1}{\epsilon}\right] \sum_{k} \Psi(\vec{k}) \ell^{ikp} = \frac{-\tau}{\epsilon_{1}} \frac{\rho}{\omega} \sum_{k} \vec{k} \Psi(\vec{k}) \ell^{ikp}$$
(12)

Before we proceed with its analysis we shall generalize this equation for the case when the dielectric constant of the film (but not of the substrate) depends on the electric field strength:

$$\epsilon_1(\omega, \vec{p}, \vec{z}) = \epsilon_1(\omega) + \propto |E(\vec{p}, \vec{z})|^2$$
(13)

RESULT AND DISCUSSION

For the above mention generalization it is reasonable to consider first of all equation (12) of the linear theory.

In this the small parameter $\tau(\vec{k})$ is present in the combination $z = \tau k/\epsilon_1$, since we analyze a region of small ϵ_1 (ω), then though $\tau k \ll 1$, the parameter z may already be generally speaking, of the order of unity. Naturally, in the nonlinear theory we are also interested only in the terms of the order of z and not τk . [3]. Since these terms appear through the use of the relationships equation (9) and the continuity of the z

component of electric displacement generalization of equation (12) is quite trivial. It turns out that in equation (12) in order to take into account nonlinearity. It is necessary to substitute $\in_1(\omega)$ with the value equation (13) at $\mathbb{Z} = 0$. Thus, when \in depends on \overrightarrow{p} (it is clear that the relationship equation (13) corresponds only to a specific case of this dependence) the equation for $\Psi(\overrightarrow{k})$ becomes:

$$\left[1 + \frac{1}{\epsilon_2}\right] \sum_{\mathbf{k}} \Psi(\mathbf{\vec{k}}) \ell^{ikp} = \frac{-\tau (\mathbf{\vec{\rho}})}{\epsilon_1 (\omega \ \mathbf{\vec{\rho}})} \sum_{\mathbf{k}} |\mathbf{\vec{k}}| \Psi(\mathbf{\vec{k}}) \ell^{ikp}$$
(14)

If nonlinearity for equation (13) is taken into account

$$\begin{array}{l} \displaystyle \in_{1}(\omega,\overrightarrow{p}) = \in_{1}(\omega) + \infty |\overrightarrow{E}(\overrightarrow{p},0)|^{2} \\ \text{and since, due to } |\in_{1}| << 1, | E_{z}^{f}| = \frac{D_{z}^{f}}{\epsilon_{1}} = \frac{D_{z}^{v}}{\epsilon_{1}} = \frac{E_{z}^{v}}{\epsilon_{1}} >> |E_{x,y}^{v}| \end{array}$$

We have

$$\epsilon_{1}(\omega, \vec{p}) = \epsilon_{1}(\omega) + \frac{\alpha}{\epsilon^{2}_{1}(\omega)} |E^{v}_{z}(\vec{p}, 0)|^{2}$$

$$\text{where } E^{v}_{z}(\vec{p}, 0) = \sum \vec{k} |\Psi(\vec{k})|^{ikp}$$

$$(15)$$

Thus, in the frequency region under discussion ($|\in_1(\omega)| << 1$) nonlinearity may be important at relatively smaller field values in vacuum since here the effective value of the nonlinearity coefficient α increase sharply instead of α there appears $\alpha = \alpha/\epsilon_1^2 (\omega), |\infty| >> |\infty|$. It may be convenient to rewrite equation (14) in such a way where its nonlinear part is written in the explicit form, if we introduce the following notations:

$$\epsilon_{1}(\omega) = \epsilon_{1}(\omega) \left[1 + \frac{1}{\epsilon_{2}} \right]^{3}$$
$$\beta = \infty \left[1 + \frac{1}{\epsilon_{2}} \right]^{3}$$

this equation may be written as follows:

0

$$\dot{\vec{\epsilon}}_{1}(\omega)\sum_{k}\Psi(\vec{k})\ell^{ikp} + \tau(\vec{\rho})\sum_{k}\vec{k}|\Psi(\vec{k})\ell^{ikp} = -\hat{L}[\Psi]$$
(16)

where

$$-\overset{\wedge}{\mathbf{L}}[\Psi] = \frac{\beta}{[\epsilon(\omega)]^2} \{ \sum_{k} |\widetilde{k}| \Psi(\widetilde{k}) \ell^{\widetilde{k}p} \}^2 \sum_{k} \Psi(\widetilde{k}) \ell^{i\widetilde{k}p}$$
(17)

We shall analyze equation (16) and try to use it to clarify possible existence of so called nonlinear local resonances i.e. those vibrations in the film polarization localized in the rough region the very existence of which is conditioned by the nonlinear term in equation (16). First of all, similarly to the case of the linear theory, we assume that $\tau = \tau$ (x) and seek the local states of all small radius r << L. Besides, in the expression equation (16) we substitute the field square by its value averaged over the local state. In this approximation equation (16) becomes the linear, i.e., it goes over into equation of the form equation (16) with L = 0; however, instead of $\in (\omega)$ there appears a new "effective" value independent of x. [4]

$$\epsilon_{1}^{NL}(\omega) = \epsilon_{1}(\omega) + \frac{\beta}{\left[\epsilon(\omega)\right]^{2}} A_{n}^{2}$$
(18)

where $A_n^2 = \int |\Psi_n(x)|^2 |E_n^v(x)|^2 dx$

 $\Psi_n(x)$ is the potential distribution in the n th local state. In this approximate the Fourier components of the function satisfy the equation β

$$\overline{\left[\in_{1}(\omega)\right]^{2}}$$

$$\in_{1}^{NL}(\omega_{n})\sum(k)\ell^{ikx} + \tau(x)\sum|k|\Psi_{n}(k)\ell^{ikx} = 0$$
(20)

In this approximate we obtain an equation for $\rho(k)$ which coincides with the Schrodinger equation for the S-state ($\ell = 0$) of the election in a hydrogen atom:

$$-\frac{1}{2n} \frac{d^2 \rho}{dQ_k^2} - \frac{\ell^2}{|Q|} \rho = E \rho_k^{\ }$$
(21)

where

$$E = -\tau(0)\eta''(0)\left[\frac{1}{\overline{\epsilon_1}^{\mathsf{NL}}(\omega)}\right]^2 = K \left|\frac{\overline{\epsilon_1}^{\mathsf{NL}}(\omega)}{\eta''(0)}\right|,$$
(22)

From the quantization condition $E = -\frac{1}{2n^2}$ we find that the frequencies of nonlinear resonance are determined by the relationship.

determined by the relationship:

$$\overline{\epsilon_1}^{\text{NL}}(\omega_n) = n\sqrt{2\tau(0)} |\eta^{"}(0)|, \quad n = 1,2,3$$

or taking into account equation (18) and (17)

$$\omega_{\rm n} - \omega_{\rm o} + \frac{\beta \overline{\Delta}^3}{\omega_{\rm n} - \omega_{\rm o}} \,A^2 = -n\Omega \tag{23}$$

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(19)

where $\Omega = \overline{\Delta}\sqrt{2\tau(0)}|\eta''(0)|$, so that $\omega_n \to \omega_o$ at large n. From the relationship, it follows first of all that the frequencies ω_n stop being equidistant as they become function of the square amplitude of vibration A^2 . In particular, at sufficiently small A^2 .

$$\omega_{n} = \omega_{o} - n\Omega - \frac{\beta \overline{\Delta}^{3} A^{2}}{\Omega^{2} n^{2}}$$
(24)

CONCLUSION

The appearance of the dependence on A^2 of the frequencies ω_o which are connected genetically with the frequencies ω_n from the linear theory is a rather evident result of the nonlinearity influence. Possible appearance of such resonance of small radii, which are absent within the framework of the linear theory, is less trivial. [6]

In the frequency region $\omega_0 < \omega < \omega_0^{NL}$ the value $\in {}^{NL}(\omega) < 0$ and it is this region that the above mentioned nonlinear resonance of small radius may be present. From equation (24) it follows that formally their number is infinitely large (at large n: $\omega_n^{NL} - \omega_0 = \frac{\overline{\Delta} |\beta| A^2}{\sqrt{n\Omega}}$. However, due to broadening only the states with small n may be of interest. In this connection, let us evaluate $|\beta|A^2$ for such a situation when $\omega_1^{NL} - \omega_0 = \frac{\overline{\Delta} |\beta| A^2}{\sqrt{n\Omega}}$.

$$|\beta| A^2 \underline{\Omega}(\Omega + \Gamma_0) \underline{\Gamma_0} \underline{\Omega} \underline{\Omega} \underline{\Omega}^2 \underline{\Omega}^2$$

and at $\Gamma_{o}\,\Omega\,0.1$ eV, $\Omega=0.4eV,\,\Delta\underline{\Omega}\,2eV$

the value $|\beta|A^2 = 5 \ge 10^{-4}$ which lies in the region of obtainable values. Thus, this evaluation shows that the appearance of nonlinear local resonance is possible.

Possible existence of nonlinear local resonances means that with regards to optical nonlinearity e.g. colours of the films or surface must change with varying intensity of incident radiation on the surface and generally speaking, these changes may be modelled. It is clear that the same resonance in the above

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conditions must contribute to Raman scattering enhancement on the surface processes of generation of harmonics at reflection etc.

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