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A rigorous description of electrodynamical and photothermal response by the matrix formalism in layered systems

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Abstract: A description of layered samples under modulated laser beam irradiation is given. With the help of the matrix formalism charge carrier wave as well as temperature wave may be calculated.

1. INTRODUCTION

In recent years, several theoretical descriptions of energy deposition, charge carrier waves, and temperature waves in photothermal phenomena have been published [1-4]. Thus, the principle processes are well understood. However, a description which includes all effects related to layered structures does not exist. The generally used exponential absorption of the laser energy does not take into account that the optical behaviour of layered systems may rapidly change with the thicknesses of the layers which are involved.

In this paper, we will give an electrodynamic description of the charge carrier and temperature waves in layered samples. For the case of the double modulation technique [5] the dependence of the conversion factor on material parameters and layer thicknesses will be discussed.

2. THE ELECTRODYNAMICS WITH CROSS EFFECTS

2.1. The incident intensity. Field ansatz

The intensity of a modulated laser beam with the intensity I , the modulation factor m , and the modulation frequency Ω , respectively, is given as the absolute value of the mean with respect to the optical frequency ω of the Poynting vector:

$$I(t) = \bar{I} (1 + m \cos(\Omega t + \Phi)) = \bar{I} + \bar{I} \cos(\Omega t + \Phi) = \langle |\vec{S}| \rangle_\omega = \sqrt{(\epsilon_0 \epsilon_r)/(\mu_0 \mu_r)} \langle \vec{E}^*(t) \vec{E}(t) \rangle_\omega \quad (1)$$

In the case of the double modulation technique [5] the beam carries the two modulation frequencies Ω_α and Ω_β with two phases. Both parts of the incident beam deposit energy into the sample (Fig.1). The modulated energy deposition causes modulated dissipative response processes as diffusing charge carrier concentration and temperature wave. Since the optical properties itself depend on temperature and charge carrier concentration one should expect the generation of response fields. Thus, the ansatz for the electric field is

$$\vec{E}(\vec{r}, t) = \left(\vec{E}_\alpha(\vec{r}) + \vec{E}_\alpha(\vec{r}) e^{i\Omega_\alpha t} + \sum_\gamma \hat{E}_\gamma(\vec{r}) e^{i\Omega_\gamma t} \right) e^{-i\omega_\alpha t} + \left(\vec{E}_\beta(\vec{r}) + \vec{E}_\beta(\vec{r}) e^{i\Omega_\beta t} + \sum_\delta \hat{E}_\delta(\vec{r}) e^{i\Omega_\delta t} \right) e^{-i\omega_\beta t} \quad (2)$$

Fig.1: Generation of response quantities X (e.g. charge carrier concentration, temperature)

2.2. The material parameters

Instead of the complex refractive index $n+ik$ one may use the relative permittivity ϵ which is $(n+ik)^2$ in the case of an unchanged permeability. The relative permittivity in every layer can be written as

$$\epsilon_r = \epsilon^0 + \sum_{\bar{X}} \left[\epsilon(\bar{X} - X_\infty) + \partial_{\bar{X}} \epsilon|_{\bar{X} - X_\infty} \sum_{\zeta} \frac{\hat{X}_\zeta e^{i\Omega_\zeta t} + \hat{X}_\zeta^* e^{-i\Omega_\zeta t}}{2} \right]; \quad \Omega_\zeta = \Omega_\alpha, \Omega_\beta \quad (3)$$

with an undisturbed part, a time-independent part due to the dc part of the intensity, and a modulated part. This modulated part carries the time dependence of the response quantity X (e.g. charge carrier concentration, temperature).

2.3. Deposition and response fields

The equations (2) and (3) are used in the Maxwell equations. Since the modulation frequency is small compared to the optical frequency the modulation may be considered as a parametric dependence. The linear response theory then leads to equations for both the energy depositing field and the response field in every layer:

$$\Delta \vec{E} + k^2 \vec{E} = 0; \quad \Delta \hat{\vec{E}} + k^2 \hat{\vec{E}} = k^2 \left[\sum \bar{\epsilon}_X / \epsilon^0 \vec{E} + \sum \tilde{\epsilon}_X / \epsilon^0 \hat{\vec{E}} \right]; \quad k^2 = \omega^2 \epsilon_0 \mu_0 \epsilon^0 \mu^0 \quad (4)$$

The solution for the energy depositing fields in every layer l (z runs from D_{l-1} to $D_{l-1} + d_l = D_l$) in the case of an excitation with a laser beam with a beam waist w can be written in terms of field amplitude vectors:

$$E_l(z) = \begin{pmatrix} 1 & 1 \end{pmatrix} \begin{pmatrix} e^{ik_l(z-D_{l-1})} & 0 \\ 0 & e^{-ik_l(z-D_{l-1})} \end{pmatrix} \begin{pmatrix} E_l^+ \\ E_l^- \end{pmatrix} e^{i\Omega t} e^{-r^2/(2w)} = \vec{N} p_l(z) \vec{E}_l e^{i\Omega t} e^{-r^2/(2w)} \quad (5)$$

The energy deposition Ξ is given by the divergence of the Poynting vector [6]. Equations (6) and (1) give

$$\Xi_{\alpha l} = \vec{E}_{\alpha l}^* p_{\alpha l}^* D_{\alpha l} p_{\alpha l} \vec{E}_{\alpha l} e^{i\Omega_\alpha t} + c.c.; \quad D_{\alpha l} = \frac{i}{4} \omega_\alpha \epsilon_0 (\epsilon_l^0 - \epsilon_l^{0*}) \begin{pmatrix} 1 & 1 \\ 1 & 1 \end{pmatrix} \quad (6)$$

for the part of the energy deposition modulated with Ω_α in the layer l (D : deposition matrix).

In the case of the response field one gets

$$\hat{E}_l(z) = \bar{N} p_l(z) \left[\hat{\bar{E}}_l + \bar{r}_l(z) \bar{\bar{E}}_l + \bar{r}_l(z) \bar{\tilde{E}}_l \right]; \quad r(z) = \begin{pmatrix} I^0(z) & I^-(z) \\ -I^+(z) & -I^0(z) \end{pmatrix} \quad (7)$$

$$I_l^0(z) = \frac{k_l}{2i} \sum_X \frac{\partial_X \epsilon}{\epsilon_l^0} \int_{D_{l-1}}^z dz' \hat{X}(z'); \quad I_l^+(z) = \frac{k_l}{2i} \sum_X \frac{\partial_X \epsilon}{\epsilon_l^0} \int_{D_{l-1}}^z dz' \hat{X}(z') e^{\pm 2ik_l(z'-D_{l-1})}$$

Since the response matrix r consists the response integrals I with a kernel that essentially is a response quantity (charge carrier concentration, temperature) the response fields contain the information on the sample. The boundary conditions for the electromagnetic field result in matrix equations for the amplitude vectors:

$$d_{l+1} \bar{\bar{E}}_{l+1} = d_l p_l(z=D_l) \bar{\bar{E}}_l; \quad d_l = \begin{pmatrix} 1 & 1 \\ f_l & -f_l \end{pmatrix}; \quad f_l = \sqrt{\epsilon_0 \epsilon_l / (\mu_0 \mu_l)} \quad (8)$$

The equation (8) establishes a procedure to calculate all field amplitude vectors for all layers beginning from the incident field at the sample surface [7]. Both the dynamic matrix d and propagation matrix a at the boundary are given by the material parameter. A similar equation results for the response field:

$$d_{l+1} \hat{\bar{E}}_{l+1} = d_l p_l(z=D_l) \left[\hat{\bar{E}}_l + \bar{r}_l(z=D_l) \bar{\bar{E}}_l + \bar{r}_l(z=D_l) \bar{\tilde{E}}_l \right] \quad (9)$$

But in contradiction to the energy depositing fields, the matrix formalism for the response field is inhomogeneous. The intensity which may be detected outside the sample contains beside the reflection two response intensity parts (M : reflection or transmission mode of measurement)

$$I_{\alpha M}(r) = f_M/2 \sum_Y \bar{E}_{\alpha M}^* e^{-r^2/(2w^2)} \hat{E}_{YM}(r) e^{iQ_Y t} + f_M/2 \sum_Y \hat{E}_{YM}^*(r) \bar{E}_{\alpha M} e^{-r^2/(2w^2)} e^{i(Q_\alpha - Q_Y)t} + c.c. \quad (10)$$

and depends on the radial distance between measurement and axis of the beam. In the case of the double modulation technique the signal at the difference frequency is measured. In (10) the response intensity can carry the the difference frequency in two cases. Either the response field itself carries the difference frequency and forms with the dc electric field the response intensity (first term) or both the response field and the ac energy depositing field mix their frequencies to the difference frequency (second term). Therefore, in the following only the generation of these two response electric fields is considered.

3. CHARGE CARRIER AND TEMPERATURE WAVES

In contradiction to the electromagnetic waves where the direction of the energy flux remains unchanged as long as the beam waist does, the deposited energy causes dissipative phenomena of energy transfer. These are usually the diffusion of generated excess charge carriers and the heat flow. Thus the Laplacian not only works in one direction as it was the case in (4,5). A general differential equation for dissipative waves of a quantity X (e.g. charge carrier concentration, temperature) can be obtained from the continuity equation:

$$\Delta \hat{X}(r, z) - r_X^2 \hat{X}(r, z) = -s_X \bar{\Xi}(z) e^{-(r^2/w^2)} - \sum_{X'} s_{XX'} \hat{X}'(r, z) \quad (11)$$

The inhomogeneity contains the modulated energy deposition and a coupling term with other response waves. This equation may be solved by the help of the Hankel transform [8] of order $\nu=0$. The solution of the transformed differential equation (11) is

$$\hat{X}(\lambda, z) = \vec{N} \cdot \vec{p}_X(z) \left\{ \hat{X}_\lambda - \vec{\Xi}_{\lambda X}(z) - \sum_{X'} \hat{X}'_{\lambda X}(z) \right\}; \quad r_{\lambda X}^2 = r_X^2 + \lambda^2; \quad \vec{p}_{Xl}(z) = \begin{pmatrix} e^{r_{\lambda X}(z-D_{l-1})} & 0 \\ 0 & e^{-r_{\lambda X}(z-D_{l-1})} \end{pmatrix} \quad (12)$$

with the energy deposition vector and the coupling vector

$$\vec{\Xi}_{\lambda X}(z) = \begin{pmatrix} +\frac{w^2 s_X}{4 r_{\lambda X}} e^{-\frac{w^2}{4} \lambda^2} \int_{D_{l-1}}^z dz' e^{-r_{\lambda X}(z'-D_{l-1})} \vec{\Xi}_\lambda(z') \\ -\frac{w^2 s_X}{4 r_{\lambda X}} e^{-\frac{w^2}{4} \lambda^2} \int_{D_{l-1}}^z dz' e^{+r_{\lambda X}(z'-D_{l-1})} \vec{\Xi}_\lambda(z') \end{pmatrix}; \quad \hat{X}'_{\lambda X}(z) = \begin{pmatrix} +\frac{s_{XX'}}{2 r_{\lambda X D_{l-1}}} \int_{D_{l-1}}^z dz' e^{-r_{\lambda X}(z'-D_{l-1})} \hat{X}'(\lambda, z') \\ -\frac{s_{XX'}}{2 r_{\lambda X D_{l-1}}} \int_{D_{l-1}}^z dz' e^{+r_{\lambda X}(z'-D_{l-1})} \hat{X}'(\lambda, z') \end{pmatrix} \quad (13)$$

The boundary conditions again result in a matrix equation for the amplitude vectors:

$$\vec{d}_{Xl+1} \vec{X}_{l+1} = \vec{d}_X \vec{p}_{Xl}(z=D_l) \left\{ \vec{X}_l - \vec{\Xi}_{\lambda Xl}(z=D_l) - \sum_{X'} \vec{X}'_{\lambda Xl}(z=D_l) \right\} \quad (14)$$

where the elements in the dynamic matrix for the response quantity $f_l = D_{Xl} f_{X\lambda l}$ contain the diffusivity D .

4. THE CONVERSION FACTOR

Since the response fields are only generated in the sample but not introduced from outside the characteristic measurement value in a double modulation excitation of a sample is the conversion factor. It is the response intensity normalized by the absorbed intensity and laser beam radius [9].

Fig.2: Conversion Factor for Al (30 nm)/SiO₂/Si

The matrix formalism for the calculation of the amplitude vectors was applied to a layered system with 30 nm Al on top of a SiO₂ layer on a Si substrate. The oxide layer thickness was varied. The result for $\lambda = 514$ nm with a beam radius of 5 μ m is shown in Fig.2. The dependence on the layer thickness is much stronger than for the reflectivity R which shows only change of about 10%.

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