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THE CALCULUS OF THE ABSORPTIVITY OF AN OXIDE-METAL SYSTEM BY POWERFUL CO_2 LASER IRRADIATION OF METALLIC SAMPLES IN AIR

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A rather simple equation is derived relating the optical absorptivity of a metal-oxide system on the oxide and metal optical constants and on the thickness of the oxide layer growing on the metallic surface during the powerful CO_2 laser irradiation in air.

1. Introduction

The formation of new compounds by powerful laser irradiation of solid surfaces (and, particularly, of the metallic ones[1-6]), are both of fundamental interest and of direct technological significance. Specifically, the oxygen incorporation and the formation of oxide films, as a result of pulsed or cw powerful laser iradiation of metals are interesting as they result from components prepared by such materials when subjected to powerful laser irradiation in air.

It is to be mentioned here that the oxide forming on the metal surface is essentially amplifying the absorptivity of the laser radiation[2,3,6-9], with a direct effect on the general efficiency of the different laser processing operations.

That is why the understanding and the appropriate description of oxide layer growth and especially of the related variation of the surface absorptivity of the sample is of great importance not only for laser technology development, but also as a new approach to the investigation of the laser surface thermochemical reactions.

We establish in this paper a rather simple formula allowing for the rapid estimation of the absorptivity of a metallic sample on the surface of which an oxide layer is forming during powerful CO_2 laser heating in air.

We focused here upon the case of CO_2 lasers as, due to the recent technological progress, they are presently the most involved in different kinds of metal processing (e. g. in cutting, welding, boring, thermal treatments of metals) and because they use metallic components (as mirrors, deflectors etc....) often placed in ambient air.

2. *Calculus of the oxide transfer matrix and of the reflection coefficient of the oxide-metal system. A general approach*

We shall consider the incident *TM* laser wave normal onto the surface of the oxide-metal system.

The continuity conditions for the amplitude of the electric field strength on the first and second interfaces are then to be written as

$$\begin{cases} E(-0) + E(0) = E(+0) + E(-x) \\ \sqrt{\frac{\varepsilon_1}{\mu_1}}\,[E(0) - E(-0)] = \sqrt{\frac{\varepsilon_2}{\mu_2}}\,[E(+0) - E(-x)] \end{cases} \tag{1a}$$

$$\begin{cases} E(+0)\,\mathrm{e}^{-ik_2x} + E(-x)\,\mathrm{e}^{ik_2x} = E(+x)\,\mathrm{e}^{-ik_3x} \\ \sqrt{\frac{\varepsilon_2}{\mu_2}}\,[E(+0)\,\mathrm{e}^{-ik_2x} - E(-x)\,\mathrm{e}^{ik_2x}] = \sqrt{\frac{\varepsilon_3}{\mu_3}}\,E(+x)\,\mathrm{e}^{-ik_3x}. \end{cases} \tag{1b}$$

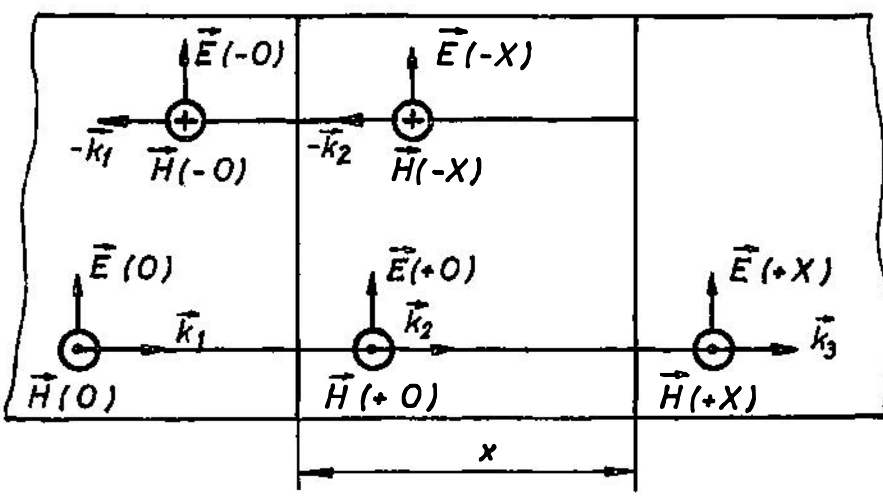

Fig. 1. Diagram showing the propagation of different electromagnetic waves into the system air-oxide-metal.

We have denoted here (Fig. 1) with $E(0)$ the amplitude of the electric field strength of the electromagnetic wave incident on the system oxide-metal; $E(-0)$, $E(+0)$ are the amplitudes of the electric field strengths of the electromagnetic waves reflected on the air-oxide boundary and of the wave passing into the oxide; $E(-x)$, $E(+x)$ are the amplitudes of the electric field strengths of the electromagnetic wave reflected on the oxide-metal boundary (and spreading into the oxide) and of the wave passing into the metal. $\vec{k}_1, \vec{k}_2, \vec{k}_3$ are the wave vectors of the incident wave, of the wave passing into the oxide and of the wave passing into the metal, respectively, while x stands for the thickness of the oxide layer forming on the metal surface.

We specify

$$\begin{cases} \vec{A}_0 = \begin{pmatrix} E(-0) + E(0) \\ \sqrt{\frac{\varepsilon_1}{\mu_1}}\,[E(0) - E(-0)] \end{pmatrix} & (2a) \\ \vec{A}_1 = \begin{pmatrix} E(+0) + E(-x) \\ \sqrt{\frac{\varepsilon_2}{\mu_2}}\,[E(+0) - E(-x)] \end{pmatrix} & (2b) \\ \vec{A}_2 = \begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_3}{\mu_3}} \end{pmatrix} E(+x)\, e^{-ik_3 x}. & (2c) \end{cases}$$

Using these notations, the first two equations of the set (1) become

$$\vec{A}_0 = \vec{A}_1 \tag{3a}$$

while the next two equations give

$$\vec{A}_2 = \begin{pmatrix} \cos k_2 x & -i\sqrt{\frac{\mu_2}{\varepsilon_2}}\sin k_2 x \\ -i\sqrt{\frac{\varepsilon_2}{\mu_2}}\sin k_2 x & \cos k_2 x \end{pmatrix} \vec{A}_1 = \hat{M}\vec{A}_1. \tag{3b}$$

Since the matrix $\hat{M}$ contains only information concerning the oxide layer growing on the metal surface we shall call it oxide transfer matrix.

From equations (3a), (3b) we get

$$\hat{M}\vec{A}_0 = \vec{A}_2. \tag{4}$$

The vectors $\vec{A}_0, \vec{A}_2$ can be further written under the form

$$\vec{A}_0 = E(0)\begin{pmatrix} 1+R \\ \sqrt{\frac{\varepsilon_1}{\mu_1}}(1-R) \end{pmatrix} = E(0)\left(\begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix} + \begin{pmatrix} 1 \\ -\sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix} R\right) \tag{5a}$$

$$\vec{A}_2 = E(0)\, T \begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_3}{\mu_3}} \end{pmatrix} e^{-ik_3 x}. \tag{5b}$$

Here we have noted with R and T the reflection coefficient and the transmission coefficient of the oxide-metal system

$$R = \frac{E(-0)}{E(0)} \tag{6a}$$

$$T = \frac{E(+x)}{E(0)}. \tag{6b}$$

Using relations (5a), (5b), equation (4) will be written in the form

$$\hat{M}\left[\begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix} + \begin{pmatrix} 1 \\ -\sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix} R\right] = \begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_3}{\mu_3}} \end{pmatrix} T\,\mathrm{e}^{-ik_3x}. \tag{7}$$

Multiplying both sides of the equation (7) with the line matrix $\left(\sqrt{\frac{\varepsilon_3}{\mu_3}}, -1\right)$, we obtain the following expression for the reflection coefficient of the oxide-metal system

$$R = \frac{\left(\sqrt{\frac{\varepsilon_3}{\mu_3}}, -1\right)\hat{M}\begin{pmatrix} 1 \\ \sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix}}{\left(\sqrt{\frac{\varepsilon_3}{\mu_3}}, -1\right)\hat{M}\begin{pmatrix} 1 \\ -\sqrt{\frac{\varepsilon_1}{\mu_1}} \end{pmatrix}}. \tag{8}$$

Introducing the Pauli matrix, $\sigma_3 = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$, and taking into account that the laser radiation is coming from the ambient air, i. e. $\varepsilon_1 \approx \mu_1 \approx 1$, we get

$$R = -\frac{\left(1, -\sqrt{\frac{\mu_3}{\varepsilon_3}}\right)\hat{M}\begin{pmatrix} 1 \\ 1 \end{pmatrix}}{\left(1, -\sqrt{\frac{\mu_3}{\varepsilon_3}}\right)\hat{M}\,\sigma_3\begin{pmatrix} 1 \\ 1 \end{pmatrix}}. \tag{9}$$

3. Calculus of the CO_2 — laser radiation absorptivity by the oxide-metal system

We shall further replace $\mu_2 \simeq 1$. Also, as the metallic oxides intensively absorb the 10.6 μm laser radiation, the wave vector and the refraction index are complex quantities and we can write[11]

$$\sqrt{\frac{\varepsilon_2}{\mu_2}} = n_2 - i\varkappa_2 \tag{10}$$

and

$$\psi = k_2 x = \frac{1}{2} \beta x - \mathrm{i} \frac{1}{2} \alpha x \tag{11}$$

where we denoted

$$\alpha = \frac{4\pi}{\lambda} \operatorname{Im} \sqrt{\varepsilon_2} \tag{12a}$$

$$\beta = \frac{4\pi}{\lambda} \operatorname{Re} \sqrt{\varepsilon_2} . \tag{12b}$$

Here n_2, $\varkappa_2$ are the refraction index and the absorption index of the oxide layer, respectively. Usually for metallic oxides at $\lambda \cong 10.6\ \mu\text{m}$, we have[11]

$$n_2 > 1, \ \varkappa_2 \ll 1 \tag{13a}$$

and so

$$\frac{\alpha}{\beta} = \frac{\varkappa_2}{n_2} \ll 1. \tag{13b}$$

Also, for most of the metals heated in air with a high power CO_2 laser radiation from the room temperature up to the metal fusion point, the metal absorptivity exhibits at most through three interferential oscillations[7–9,11,12], i. e. the oxide keeps optical thin. Therefore

$$\alpha x \ll 1. \tag{14}$$

We can write then

$$\sqrt{\frac{\mu_2}{\varepsilon_2}} \cong \frac{1}{n_2} + \mathrm{i} \frac{\varkappa_2}{n_2^2}$$

and

$$\begin{cases} \sin \psi \simeq \sin \dfrac{\beta x}{2} - \mathrm{i} \dfrac{\alpha x}{2} \cos \dfrac{\beta x}{2} & (15a) \\ \cos \psi \cong \cos \dfrac{\beta x}{2} + \mathrm{i} \dfrac{\alpha x}{2} \sin \dfrac{\beta x}{2}. & (15b) \end{cases}$$

Taking into account the equations (14), (15a), (16b) one can split the transfer matrix, $\hat{M}$ (Eq. (3b)) into two parts

$$\hat{M} = \hat{M}_0 + \hat{M}_1 \tag{16a}$$

where

$$\hat{M}_0 = \begin{bmatrix} \cos \dfrac{\beta x}{2} & - \mathrm{i} \dfrac{1}{n_2} \sin \dfrac{\beta x}{2} \\ - \mathrm{i} n_2 \sin \dfrac{\beta x}{2} & \cos \dfrac{\beta x}{2} \end{bmatrix} \tag{16b}$$

stands for the zero-order transfer matrix, while

$$\hat{M}_1 \simeq \begin{bmatrix} i\frac{\alpha x}{2}\sin\frac{\beta x}{2} & \frac{\varkappa_2}{n_2^2}\sin\frac{\beta x}{2} - \frac{\alpha x}{2n_2}\cos\frac{\beta x}{2} \\ -n_2\frac{\alpha x}{2}\cos\frac{\beta x}{2} - \varkappa_2 - \varkappa_2\sin\frac{\beta x}{2} & i\frac{\alpha x}{2}\sin\frac{\beta x}{2} \end{bmatrix} \tag{16c}$$

is a matrix comprising terms much lower than the unity and therefore will be further treated as a first order approximation matrix.

By further using the above introduced approximations, the reflection coefficient, R, can be also written in the form

$$R = R_0 + R_1. \tag{17}$$

To this purpose we first mention that if noting with Z_3 the surface impedance of the metal, we have[8-12]

$$\sqrt{\frac{\mu_3}{\varepsilon_3}} \simeq \frac{c Z_3}{4\pi} \simeq \frac{A_0}{4}(1 + i). \tag{18}$$

Then, as the absorptivity of the radiation of $\lambda \simeq 10.6\,\mu m$ by metals is very small

$$A_0 \ll 1, \tag{19}$$

we can split the line matrix

$$\vec{b} \equiv \left(1, -\sqrt{\frac{\mu_3}{\varepsilon_3}}\right) \tag{20}$$

into two terms

$$\vec{b} = \vec{b}_0 + \vec{b}_1 \tag{21a}$$

where

$$\vec{b}_0 = (1, 0) \tag{21b}$$

is a zero-order term, while

$$\vec{b}_1 = (0, 1)\left[-\frac{A_0}{4}(1 + i)\right] \tag{21c}$$

is a correction term, containing only quantities much smaller than the unity.

The reflection coefficient (9) is then to be written further as

$$R = -\frac{(\vec{b}_0 + \vec{b}_1)(\hat{M}_0 + \hat{M}_1)\vec{a}_0}{(\vec{b}_0 + \vec{b}_1)(\hat{M}_0 + \hat{M}_1)\sigma_3\vec{a}_0} \tag{22}$$

where $a_0 = \begin{pmatrix} 1 \\ 1 \end{pmatrix}$. Splitting now the reflection coefficient as in equation (17), we have

$$R_0 = -\frac{(\vec{b}_0\,\hat{M}_0\,\vec{a}_0)}{(\vec{b}_0\,\hat{M}_0\,\sigma_3\,\vec{a}_0)} \tag{23a}$$

and

$$R_1 = -\frac{1}{(\vec{b}_0\,\hat{M}_0\,\sigma_3\,\vec{a}_0)}\,[(\vec{b}_1\,\hat{M}_0\,\vec{a}_0) + (\vec{b}_0\,\hat{M}_1\,\vec{a}_0)] + \tag{23b}$$

$$+\frac{(\vec{b}_0\,\hat{M}_0\,\vec{a}_0)}{[(\vec{b}_0\,\hat{M}_0\,\sigma_3\,\vec{a}_0)]^2}\,[(\vec{b}_1\,\hat{M}_0\,\sigma_3\,\vec{a}_0) + (\vec{b}_0\,\hat{M}_1\,\sigma_3\,\vec{a}_0)].$$

The complex conjugated, zero-order reflection coefficient will be equal to

$$R_0^* = -\frac{(1,0)\,\sigma_3\,\hat{M}_0\,\sigma_3 \begin{pmatrix} 1 \\ 1 \end{pmatrix}}{(1,0)\,\sigma_3\,\hat{M}_0\,\sigma_3 \begin{pmatrix} 1 \\ 1 \end{pmatrix}}. \tag{24}$$

Here we have taken into account that

$$\hat{M}_0^* = \sigma_3\,\hat{M}_0\,\sigma_3. \tag{25}$$

As

$$\sigma_3^2 = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix} \tag{26a}$$

$$(1,0)\,\sigma_3 = (1,0) \tag{26b}$$

we get

$$|R_0|^2 = 1. \tag{27}$$

From

$$A = 1 - |R|^2 \tag{28}$$

we obtain then

$$A \simeq -2\,\mathrm{Re}\,(R_0^*\,R_1) \tag{29}$$

or taking into account relations (23b) and (24) we have

$$A = \frac{1}{(\vec{b}_0\,\hat{M}_0\,\vec{a}_0)}\,[-(\vec{b}_1\,\hat{M}_0\,\vec{a}_0) + (\vec{b}_1^*\,\sigma_3\,\hat{M}_0\,\vec{a}_0) -$$

$$-(\vec{b}_0\,\hat{M}_1\,\vec{a}_0) - (\vec{b}_0\,\sigma_3\,\hat{M}_1\,\vec{a}_0)] + \frac{1}{(\vec{b}_0\,\hat{M}_0\,\sigma_3\,\vec{a}_0)}\cdot \tag{30}$$

$$[-(\vec{b}_1^*\,\sigma_3\,\hat{M}_0\,\sigma_3\,\vec{a}_0) + (\vec{b}_1\,\hat{M}_0\,\sigma_3\,\vec{a}_0) + (\vec{b}_0\,\sigma_3\,\hat{M}_1\,\sigma_3\,\vec{a}_0) + (\vec{b}_0\,\hat{M}_1\,\sigma_3\,\vec{a}_0)].$$

Using expressions (21b), (21c) of the line matrices $\vec{b}_0, \vec{b}_1$ one gets

$$A = \frac{1}{(\vec{b}_0\, \hat{M}_0\, \vec{a}_0)} \left[- \left(0, -\frac{A_0}{2}\right) \hat{M}_0 \begin{pmatrix} 1 \\ 1 \end{pmatrix} - 2\,(1, 0)\, \hat{M}_1 \begin{pmatrix} 1 \\ 1 \end{pmatrix} \right] +$$

$$+ \frac{1}{(\vec{b}_0\, \hat{M}_0\, \sigma_3\, \vec{a}_0)} \left[2(1, 0)\, \hat{M}_1\, \sigma_3 \begin{pmatrix} 1 \\ 1 \end{pmatrix} - \left(0, \frac{A_0}{2}\right) \hat{M}_0 \sigma_3 \begin{pmatrix} 1 \\ 1 \end{pmatrix} \right]. \quad (31)$$

Finally, using Eqs. (16b) and (16c), we obtain after usual calculations

$$A = \frac{n_2^2\, A_0 + 2\, \varkappa_2\, (\beta\, x - \sin \beta\, x)}{n_2^2 + (1 - n_2^2) \left(\sin^2 \frac{\beta\, x}{2}\right)}. \quad (32)$$

One can use the rather simple equation (32) in order to estimate (i) the absorptivity of a metal-oxide system under powerful CO_2 laser irradiation in air or (ii) to characterize the growing kinetics, $x\,(t)$ on the surface of the metallic sample during the powerful CO_2 laser irradiation in air. To this purpose determinations of the thickness of the oxide layer at different laser dwelling times or time resolved absorptivity measurements, $A\,(t)$ (for details see Refs. 1,3,7 and 9), could be performed.

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RAČUN MOĆI APSORPCIJE SISTEMA METALNIH OKSIDA KOD SNAŽNOG ZRAČENJA CO_2 LASEROM NA METALNIM UZORCIMA U ZRAKU

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Izvedena je dosta jednostavna jednadžba za optičku moć apsorpcije sistema metal-oksid u zavisnosti od optičkih konstanata oksida i metala i od debljine sloja oksida koji nastaje na metalnoj površini za vrijeme snažnog zračenja CO_2 laserom u zraku.