Higher harmonic generation at metal surfaces by powerful femtosecond laser pulses

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We present a simple model calculation to interpret the recent data of Kohlweyer *et al.* [Opt. Commun. **117**, 431 (1995)] and von der Linde *et al.* [Phys. Rev. A **52**, R25 (1995)] on the generation of higher harmonics at metal surfaces by powerful femtosecond laser pulses. The model presents an adaptation of our earlier work [Phys. Rev. A **49**, 3106 (1994)] to the present situation. [S1050-2947(96)01210-3]

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I. INTRODUCTION

The generation of harmonics by irradiating atoms with a powerful laser beam has become a topic of increasing research interest. Summaries on this subject can be found in the book by Gavrila [1] and in the reviews by Wahlström [2] and Balcou *et al.* [3]. Since the density of electrons at the surface of a solid (a metal, in particular) is much larger than the one usually available in a beam of atoms, it is of equal interest to investigate the generation of harmonics by shining laser light on a solid surface, in particular, metal.

If the laser power is sufficiently low, perturbation theory in powers of the electric-field strength can be applied and the generation of the second harmonic will be dominant. Details on this topic can be found in the book by Shen [4]. More recent investigations, based on the jellium model of a solid, are those of Murphy *et al.* [5] and Liebsch and Schaich [6] where further references are quoted. As these investigations show, the maximum efficiency of harmonic production has to be expected if the laser light polarization is perpendicular to the solid surface and a similar result holds for the polarization of the harmonic light. At intermediate intensities of a Nd:YAG laser of about 2 GW power output, Farkas *et al.* [7] observed a few years ago the production of harmonics up to the fifth order by irradiating a gold surface at grazing incidence to have the electric-field component perpendicular to the metal surface sufficiently large. Moreover, to prevent the formation of a surface plasma, picosecond pulses were used. At about the same time two model calculations appeared addressing the same problem. In the papers by Mishra and Gersten [8,9] the metal is described by Sommerfeld's free-electron model [10] and the harmonic production rates are evaluated by means of perturbation theory in powers of the electric-field strength. Hüller and Meyer-ter-Vehn [11], on the other hand, suggest in their work a classical model which takes into account the lattice structure of the solid and the electron correlations. The production rates, predicted by these two models, are, however, not in agreement with the experimental results of Farkas *et al.* [7]. Stimulated by these investigations, we performed a simple model calculation of harmonic generation at a metal surface [12]. On the one hand it was based on Sommerfeld's model for a metal and on the other hand we applied the Kramers-Henneberger transformation [13,14] to the Schrödinger equation of the problem. The rates of harmonic generation, evaluated in this investigation, agreed quite well with the findings of Farkas *et al.* [7].

The situation is rather different if the laser power is so high that a plasma is formed at the surface of the solid by the radiation pulse. About fifteen years ago, the first experiments were performed by Carman and co-workers [15,16] yielding multiharmonic generation extending far into the UV region. These authors used a CO_2 laser of 10^{14} to 10^{16} Wcm^{-2} power output and nanosecond pulse duration. Harmonics up to the 46th order were observed by irradiating plastic- or metal-coated plane- or microballoon targets. The observed harmonic production rates showed a plateau up to a cutoff at rather high order. The theoretical interpretation of these results suggested by Bezzerides, Jones, and Forslund [17] and Grebogi, Tripathi, and Chen [18] is based on classical plasma considerations, assuming that the surface has a sharp density profile such that the electrons perform a highly nonlinear oscillation in the laser field. In these experiments, however, rather long laser pulses were used and, hence, the thickness of the plasma layer turned out to be appreciably larger than the laser wave length, since during the pulse duration the plasma had time to expand.

In more recent experiments, a second-order harmonic generation was observed by Engers *et al.* [19] and von der Linde *et al.* [20] by irradiating metal films and glass targets with femtosecond (fs) laser pulses of some 10^{16} to 10^{17} Wcm^{-2} of a mode-locked dye laser operating at 620 nm. Here the hydrodynamic expansion of the laser produced surface plasma during the much shorter interaction time is not significant. A very thin layer of high temperature and high plasma density gets generated at the surface of the solid. The extremely steep plasma density gradient is a distinctive feature of plasmas generated by fs laser pulses. Even more interesting to us will be the very recent experiments by Kohlweyer *et al.* [21] and von der Linde *et al.* [22] in which aluminum and glass targets were irradiated at intensities of 10^{17} Wcm^{-2} using a tabletop Ti:sapphire terawatt laser system yielding fs laser pulses of 800 nm wave length. In the first of these experiments harmonics up to the seventh order were observed, while in the second investigation harmonics up to the fifteenth order were detected. Here too, the very

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steep density profile of the surface plasma is essential for the production of higher harmonics.

II. THEORY OF HARMONIC PRODUCTION

After these preliminaries, it will be the purpose of the present work to show that with slight modifications of our simple quantum-mechanical model [12], mentioned above, we are able to describe quite well some of the basic features of the experimental data of Kohlweyer *et al.* [21] and von der Linde *et al.* [22]. As in Sommerfeld's model [10], we describe the laser heated surface of a metal by a potential step, the height of which is given by $V_o = E_F + W$ where E_F is the Fermi energy and W the work function of the metal. The laser polarization $\boldsymbol{\varepsilon} = \mathbf{e}_z$ is taken perpendicular to this surface and along the z direction. Since the plasma layer on the surface of the metal is very thin due to the strong radiation pressure excerted by the powerful radiation field, its electron density is about the same as that of the bulk metal [22]. Electrons of the metal scatter at the potential wall in the laser field within an interaction region having a dimension less than a wavelength along the metal surface. During this scattering process harmonics of the laser light are generated, as we have shown in a recent paper [23]. Going over to the Kramers-Henneberger frame of reference [13,14], the scattering of electrons from within the surface layer at the potential wall, which is now periodically oscillating in time, represents a highly nonlinear oscillating system the Fourier components of which will be evaluated in the following.

If we write down the Schrödinger equation for an electron moving in the potential $V(\mathbf{x})$ and simultaneously in the laser field, as well as in the effective spontaneously emitted field, then we obtain by performing the Kramers-Henneberger transformation and by considering only single-quantum spontaneous emission the following effective radiation potential [12,24]:

$$V_{\alpha,\alpha'} = \boldsymbol{\alpha}'(t) \cdot \boldsymbol{\nabla} V(\mathbf{x} + \boldsymbol{\alpha}(t)) + \cdots \tag{1}$$

with

$$\boldsymbol{\alpha}'(t) = \alpha_o' \boldsymbol{\varepsilon}' \sin \omega' t, \quad \alpha_o' = eF_o'/m(\omega')^2,$$

$$F_o' = 2(2\pi\hbar\omega'/L^3)^{1/2}, \tag{2}$$

$$\boldsymbol{\alpha}(t) = \alpha_o \boldsymbol{\varepsilon} \sin \omega t, \quad \alpha_o = eF_o/m\omega^2, \tag{3}$$

where $\boldsymbol{\varepsilon}$, ω, and F_0 are the polarization, frequency, and field amplitude of the laser radiation in dipole approximation, respectivity, and the analogous primed quantities refer to the spontaneously emitted field. By performing now first-order time-dependent perturbation theory in terms of the eigenstates of the Hamiltonian in the absence of the fields, we get the transition amplitudes

$$c_{fi}^{(1)}(t\to\infty) = -(i/\hbar)\int_{-\infty}^{+\infty} \langle f|\alpha'(t)\cdot\nabla V \times (\mathbf{x}+\boldsymbol{\alpha}(t))|i\rangle \exp(i\omega_{fi}t)\,dt \tag{4}$$

and using the Fourier expansion

$$V(\mathbf{x}+\boldsymbol{\alpha}(t)) = \sum_n V_n(\mathbf{x})\exp(-in\omega t) \tag{5}$$

we obtain the transition matrix elements of harmonic generation

$$T_{fi} = \sum_n T_n,$$

$$T_n = -2\pi i\,\delta(E_f + \hbar\omega' - E_i - n\hbar\omega)(\alpha_o'/2i) \times \langle f|\boldsymbol{\varepsilon}'\cdot\boldsymbol{\nabla} V_n(\mathbf{x})|i\rangle \tag{6}$$

In our particular case, $|i\rangle$ and $|f\rangle$ will essentially be free electron states $|\mathbf{k}\rangle$ and $|\mathbf{k}'\rangle$ of wave vectors $\mathbf{k}$ and $\mathbf{k}'$, respectively, with the corresponding free particle energies $E = (\hbar k)^2/2m$ and $E' = (\hbar k')^2/2m$. Since we have chosen the laser polarization $\boldsymbol{\varepsilon} = \mathbf{e}_z$ perpendicular to the solid surface, oriented in the (x,y) plane, the oscillating step potential will take the form

$$V(z + \alpha_o \sin\omega t) = V_o[\theta(z + \alpha_o \sin\omega t) - 1], \tag{7}$$

where $\theta(z)$ is the step function. Hence, the gradient of (7), being a δ function, will yield after Fourier decomposition

$$\nabla_z V(z + \alpha_o \sin\,\omega t) = \sum_n \nabla_z V_n(z)\exp(-in\omega t),$$

$$\nabla_z V_n(z) = V_o(2\pi)^{-1}\int dk\,\exp(-ikz)J_n(k\alpha_o). \tag{8}$$

The reflection of the electrons from the potential wall, representing the plasma layer, we shall treat in the Born approximation by neglecting the usual boundary conditions at the wall and by representing the ingoing and outgoing electrons by plane waves in which case we have

$$\langle \mathbf{x}|\mathbf{k}\rangle = L^{-1/2}\exp(ik_z z)L^{-2}\exp(i\mathbf{k}_\parallel\cdot\mathbf{x}_\parallel) \tag{9}$$

and a similar expression holds for $\langle \mathbf{x}|\mathbf{k}'\rangle$. $\mathbf{k}_\parallel$ and $\mathbf{x}_\parallel$ in (9) denote the corresponding vector components in the (x,y) plane. With this simplification we find for the matrix elements in (6)

$$\boldsymbol{\varepsilon}\cdot\langle \mathbf{k}'|\nabla_z V_n(z)|\mathbf{k}\rangle = \boldsymbol{\varepsilon}'\cdot\boldsymbol{\varepsilon} V_o L^{-1} J_n((k_z - k_z')\alpha_o)\delta_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel}, \tag{10}$$

where the Kronecker δ reflects the fact that the electron momenta parallel to the solid surface are constants of motion.

Now the total transition probabilities per unit time and unit surface are the properly weighted sums over the allowed electron and photon states [12] of the probabilities $|T_n|^2 T^{-1}$ evaluated from (6) and (10)

$$w_n = \sum_\kappa \sum_{k'} \sum_k 2[1 - f(\mathbf{k}')]2f(\mathbf{k})|T_n|^2 T^{-1} L^{-2}, \tag{11}$$

where $\kappa = \omega'/c$ and $f(\mathbf{k})$ is the Fermi-distribution function. After having changed in (11) from summations to integra-

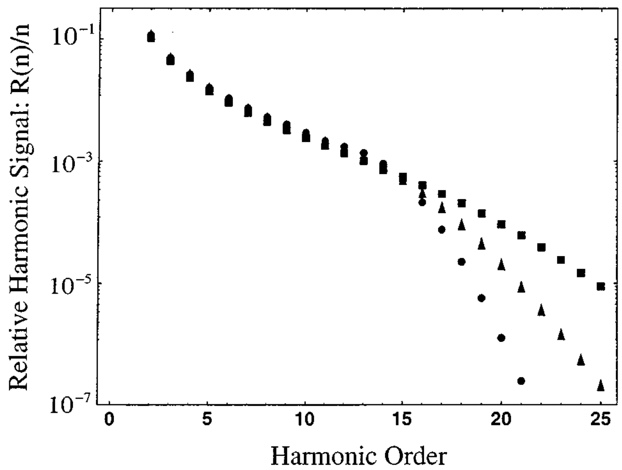

FIG. 1. The harmonic rates normalized to the fourth harmonic are shown on a logarithmic scale as a function of the harmonic order n for the three different temperatures considered and based on the parameter values of von der Linde *et al.* [22]. With increasing temperature more higher harmonics are produced.

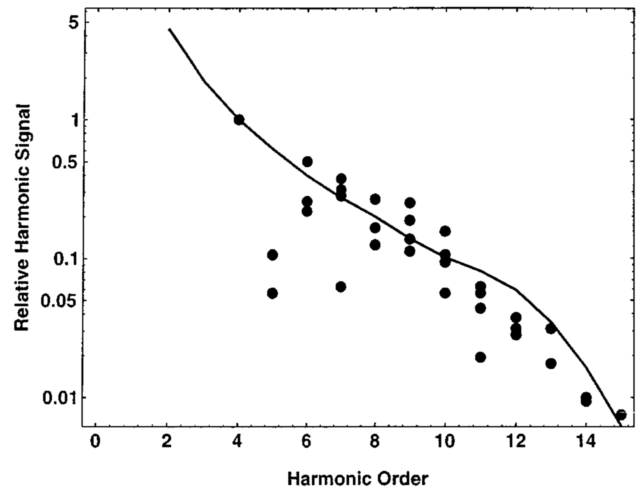

FIG. 2. Depicts on a similar scale as in Fig. 1 the experimental data of von der Linde *et al.* [22] (their Fig. 3) and shows our theoretical rates normalized to the fourth harmonic. In the range of harmonics observed, the theoretical data for all three temperatures coincide.

tions in the usual way, we use the following approximations in order to be able to perform most of the integrations analytically. We find

$$[1-f(E+\Delta E)]f(E)\simeq[1-f(E)]f(E)\,\delta(\Delta E/k_BT) \tag{12}$$

and

$$[1-f(E)]f(E)\simeq-2mk_BT\hbar^{-2}\partial f(E)/\partial(k_\parallel^2), \tag{13}$$

where $\Delta E=E_z'-E_z=n\hbar\omega-\hbar\omega'$ on account of the δ function in $|T_n|^2$. Having performed the above integrations, we get for the differential production rates per unit surface element of harmonics of frequency $\omega'=n\omega$ (n=2,3,4,...)

$$dw_n/d\Omega=[32/(2\pi)^4](e^2/\hbar c)[(V_ok_BT)^2/(\hbar c)^2\hbar^2\omega]\times(\boldsymbol{\epsilon}'\cdot\boldsymbol{\epsilon})^2R_n/n, \tag{14}$$

where R_n, expressed in dimensionless parameters, reads

$$R_n=\int_o^\infty(dy/y)\{\exp[b(y^2-1)]+1\}^{-1}J_n^2(ay), \tag{15}$$

with

$$a=2k_F\alpha_o=2k_FeF_o/m\omega^2,\quad b=E_F/k_BT, \tag{16}$$

where $(\hbar k_F)^2/2m=E_F$, the Fermi energy. Hence, according to (14), the shape of the spectrum of harmonics is governed by R_n/n. In the figures, shown below, we shall present relative rates for comparison with the experimental data. For evaluating the absolute rates from (14) we need the explicit value of the prefactor which is given by 5.847×10^{20} $[V_ok_BT\ (\text{eV})]^2$ $[\hbar\omega(\text{eV})]^{-1}$ cm^{-2} sec^{-1}, expressing the relevant values of V_o, T, and ω in electronvolts. If in (15) the parameter b is much larger than unity then the Fermi distribution has a sharp drop at $y=1$. On the other hand, $J_n^2(ay)$ gets very small as soon as the index n gets larger than the argument ay. Hence, we expect a cutoff of the harmonic rates at $n_c=a$. As the figures, shown below, indicate, this is actually the case. There is another point which has to be considered. In the definition of the parameter a in (16), F_o is the normal component of the electric-field strength within the plasma layer. Hence, in order to fulfill the boundary conditions of electrodynamics $F_o=F_a\epsilon^{-1}$ where F_a is the normal component of the impinging laser field and $\epsilon=1-(\omega_p/\omega)^2$ is the dielectric constant of the plasma, if collisions are neglected. In fact, for the temperatures considered below $\omega\tau\gg1$ where τ is the average time between two collisions. Since in the present case $\omega_p\gg\omega$ the external field component F_a will have to be reduced by a factor $(\omega/\omega_p)^2$ in order to yield the effective field inside the plasma layer. Moreover, because of the superposition of the incoming and reflected fundamental laser field, the parameter a has to be multiplied by a factor $2\sin\theta$, where θ is the angle of incidence which is 45° in the work of Kohlweyer *et al.* [21] and 68° in the experiments of von der Linde *et al.* [22].

III. COMPARISON WITH EXPERIMENTS

For a comparison of our theory with experiments, we consider the data of the harmonic rates obtained by Kohlweyer *et al.* [21] and von der Linde *et al.* [22] for aluminum targets. (Unfortunately, our model cannot be applied to nonmetallic materials.) For their experiments these authors used Ti:sapphire lasers with wave length λ=800 nm, corresponding to photon energies $\hbar\omega$=1.56 eV and frequencies $\omega=2.5\times10^{15}$ sec^{-1}. The peak power output was $I=10^{17}$ Wcm^{-2} and the pulse duration of a few hundred femtoseconds. For the Al target we find from the tables in the book of Ashcroft and Mermin [25] for the Fermi energy E_F=11.7 eV and for the work function W=4.25 eV, so that the depth of the potential well of Sommerfeld's free-electron model of the metal is V_0=15.95 eV. From the same tables we get for the electron density in Al, $n=18.1\times10^{22}$ cm^{-3} which roughly also corresponds to the electron density in the plasma layer and yields a plasma frequency $\omega_p=2.41\times10^{16}$

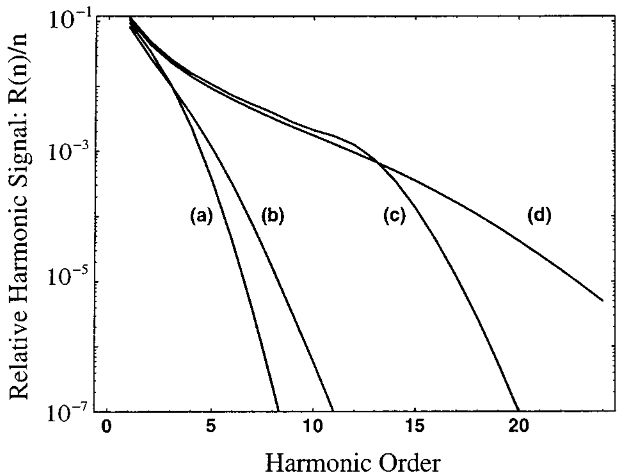

FIG. 3. Shows the development of a plateau of the harmonic rates with increasing temperature and intensity of the surface plasma and laser beam intensity as explained in the text.

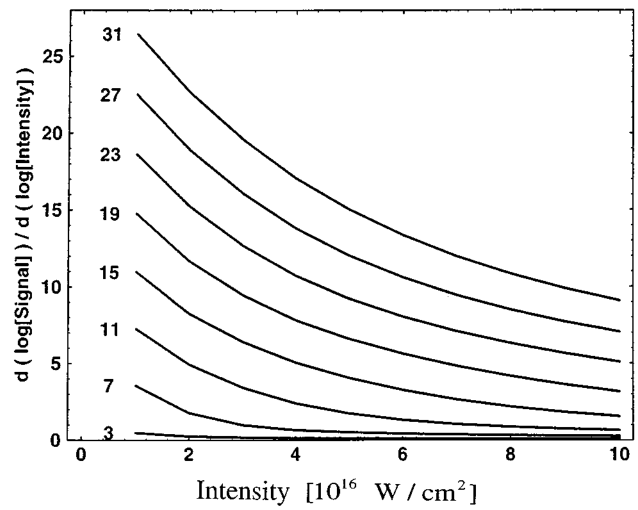

FIG. 4. Shows the degree of nonlinearity of the harmonic signal as a function of the laser field intensity. In the intensity range considered, perturbation theory in powers of the laser field intensity does not apply but the tunneling regime has not yet been reached.

$\sec^{-1}$ or $\hbar\omega_p = 15.82$ eV. We shall evaluate the harmonic spectra R_n/n of (14) in relative units for three different temperatures, namely, $T = 300$ K with $k_B T = 0.025$ eV, $T = 2.4\times 10^4$ K with $k_B T = 2$ eV, and $T = 6\times 10^4$ K with $k_B T = 5$ eV. The last two temperature values correspond to an A1-surface plasma generated by a fs laser pulse, as discussed in the work of Mancini *et al.* [26]. Thus we get for the parameter b, defined in (16), the respective values $b = E_F/k_B T = 468$, 5.58, and 2.34. Similarly, we find for the ''effective'' parameter $a_1 = 2a(\omega/\omega_p)^2 \sin\theta$, to be used in (15) instead of a, by taking the data presented above and the definition (16) for a, the following values for the two experiments above $a_1 = 16.015$ and $a_1 = 12.215$, respectively. All the figures presented below, are based on these parameter values for b and a_1.

In Fig. 1 we show the harmonic spectra $(R_n/n)/(R_4/4)$, i.e., normalized to the forth harmonic, as a function of the harmonic order n for the parameters of the experiment of von der Linde *et al.* [22] for the three temperatures mentioned before. As we can see, up to $n \simeq 15$ all three spectra essentially coincide, but with increasing temperature increasingly higher harmonics are produced. In the figure, points correspond to $T = 300$ K, triangles to $T = 2.4\times 10^4$ K and squares refer to $T = 6\times 10^4$ K.

In order to make the comparison of the predicted harmonic rates of our theory with the experimental data for A1 of von der Linde *et al.* [22] more transparent, we plotted in Fig. 2 the experimental rates as points and compare them with our results for R_n/n, normalized to the experimental rate for $n = 4$, drawn as a continuous line. The overall agreement is surprisingly good, except for the experimental rate for $n = 5$. This agreement holds for all three temperatures discussed in Fig. 1.

At this point we would like to remark that in general for $2 \leqslant n \ll a_1$ the integral in (15) can be well approximated by (see Gradshteyn and Ryzhik [27])

$$\int_0^\infty dx J_n^2(x)/x = 1/2n, \qquad (17)$$

so that the n dependence of the harmonic signal (14) is governed by the simple function $1/2n^2$. Hence, the ratio of the consecutive harmonic signals can be approximated by $n^2/(n+1)^2$, which is practically equal to one for large n. Consequently, for very large intensities I of the laser pulse (i.e., large effective a_1), a broad plateau will show up in the harmonic spectrum. Such a plateau formation was found in the early experiments by Carman, Rhodes, and Benjamin [16] by irradiating metal coated pellets with a powerful CO_2-laser beam of $I = 10^{16}$ Wcm^{-2}.

As an illustration for the plateau formation, we present in Fig. 3 for the parameters a_1 evaluated for the two laser intensities $I = 10^{16}$ Wcm^{-2} and $I = 10^{17}$ Wcm^{-2} and b calculated for the two temperatures $T = 300$ K and $T = 6\times 10^4$ K the corresponding harmonic rates. In this figure, the data presented in (a) and (b) for the relative rates $(R_n/n)/(R_4/4)$ refer to the lower laser intensity and the data of (c) and (d) belong to the higher intensity. As we can recognize, at the lower intensity the plateau of the harmonic rates is not yet strongly pronounced by going from the lower to the higher temperature [i.e., (a) to (b)], whereas at the higher laser intensity the plateau becomes apparent, in particular, by going from (c) to (d).

In Fig. 4 we present the variation of the slopes of the derivative $d\log(R_n/n)/d\log I$ for $n = 3,7,...31$ as a function of the intensity. If the production of harmonics would be in the regime of perturbation theory, we would get horizontal lines for the appropriate n value marked along the ordinate axis. As we see, we are far from this regime at the intensities considered. The evaluated slopes are always considerably smaller than n, but they are not negative so that in this intensity range we are not yet in the tunneling regime but in the transition region.

The data of the experiments of Kohlweyer *et al.* [21] are not as conveniently accessible to theoretical interpretation as the results of von der Linde *et al.* [22]. Nevertheless, we tried to compare their data for the intensity dependence of the third-harmonic rates with our predictions. This is shown

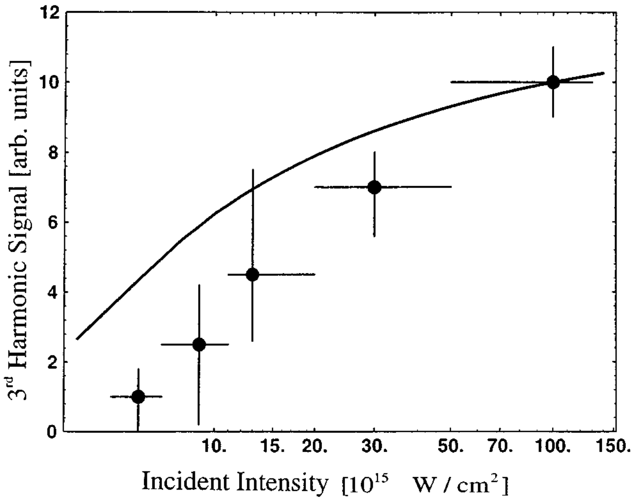

FIG. 5. Considers the intensity dependence of the third harmonic in the experiments of Kohlweyer *et al.* [21] (their Fig. 5) and makes a comparison with our theoretically predicted rates normalized to the experimental value at the highest intensity.

in Fig. 5. Here the experimental data are taken, including the error bars, from Fig. 5 of Kohlweyer *et al.* [21] and our theoretical values for the intensity dependence of $R_3/3$ are normalized to the experimental result at the highest intensity considered. The scales of our figure have been chosen in agreement with the experimental work. The overall agreement of our theoretical data with the experimental values is reasonable though our intensity dependence is less pronounced.

IV. CONCLUSIONS

Summarizing we may say that, despite the apparent simplicity of our model, we get a surprisingly good overall agreement of our theoretical rates for the harmonic production at an aluminum surface irradiated by a powerful femtosecond laser pulse if compared with the data of the experiments of Kohlweyer *et al.* [21] and von der Linde *et al.* [22]. Although our model does not take into account all the complicated plasma phenomena taking place at the metal surface during the irradiation with a powerful fs laser pulse, it appears that our highly nonlinear oscillator model describes some of the essential features of the process of harmonic production. Certainly, for a full understanding of the effect more theoretical and experimental work will have to be done.

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