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Generation of harmonics during scattering of laser radiation from a metal surface having an artificially large work function

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Abstract. We consider the generation of harmonics during the scattering of laser radiation from a metal surface. The metal will be described by Sommerfeld's free-electron model and we artificially assume the metal to have an infinitely large work function. Contrary to what has been observed for a real metal, in our model the evaluated harmonic rates show the formation of a plateau, if plotted as a function of the harmonic order n. Apparently, the depth of the potential well has some influence on the plateau-formation.

1. Introduction

The generation of higher harmonics by irradiating atoms with powerful lasers has become a field of very active research. Surveys can be found in the book of Gavrila [1] and in the reviews by Eberly *et al* [2], Mainfray and Manus [3], L'Huillier *et al* [4] and Burnett *et al* [5]. Two particular features have been observed in this process: (i) the formation of a plateau and (ii) the existence of a cut-off frequency of the harmonics. Very recent papers on these phenomena are those by L'Huillier and Balcou [6], Macklin *et al* [7], Krause *et al* [8] and L'Huillier *et al* [9].

In a previous work [10] we considered harmonic production from a metal surface, describing the metal by the Sommerfeld free-electron model [11]. We treated the Schrödinger equation of this problem in the Kramers–Henneberger frame of reference [12, 13] and derived a simple expression for the harmonic production rates, taking the laser polarization perpendicular to the metal surface. This rather elementary theory permitted us to reproduce quite successfully the experimental data of harmonic generation from a gold surface obtained by Farkas *et al* [14], who used ps-pulses of a Nd:YAG laser ($\hbar\omega = 1.17$ eV) of some 2–5 $\times$ 10^9 W cm^{-2} intensity output. In this experiment, the highest harmonic produced turned out to be the 5th corresponding to a photon energy $\hbar\omega' = 5.85$ eV, which is close to the work function of gold, $W = 4.6792$ eV [14]. This cut-off behaviour agrees quite well with the formula of Krause *et al* [8], according to which $\hbar\omega'_{\max} \approx I + 3U_p$. In our case, the ionization energy I has to be replaced by the work function W and the ponderomotive energy U_p is very small at the above laser field intensity. In the mean time this cut-off behaviour of the harmonics produced at metal surfaces has been confirmed by Farkas and co-workers in a series of new experiments with different metals and laser frequencies but even higher field intensities, taking fs laser pulses in order to prevent the formation of a surface plasma [15].

In all these experiments, however, no indication of a plateau-formation of the harmonic rates, plotted as a function of the harmonic order n, was found. Therefore, we had the

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idea to investigate what happens if we artificially increase the work function of the metal (taking the extreme case $W \to \infty$ for simplifying the treatment) and consider at the same time higher laser field intensities, in order to somehow mimic the situation of harmonic generation from an atom having a large ionization energy, and, consequently, having the ionization probability strongly suppressed. As we shall show in the present work, in this case a plateau actually appears.

In this somewhat artificial model calculation we keep in mind that the laser field, while interacting with the metal surface, penetrates within a thin layer into the metal. Here the field can only interact with the electrons during their reflection from this particular wall of the potential well to which the Fermi-gas of quasi-free electrons is bound. During this reflection process only, the electrons can emit radiation spontaneously. Since in the Sommerfeld model the potential well is essentially assumed to be of macroscopic size, it will be sufficient, to consider only the reflection process in the laser field from this one particular (infinitely high) potential wall. As we have discussed in our earlier work [10], only those electrons will essentially be involved in the reflection process, whose energy E is close the the Fermi-energy E_F. During the electron reflection in the laser field the electrons can be excited to energies $E_n = E + n\hbar\omega$ by absorbing n photons from the field and, at the same time can fall back to a state of energy E' which is again close to E_F. In the ideal case, apparently realized experimentally [14], we find $E' = E = E_F$, which will be true, in particular, at absolute temperature $T = 0$ [10]. Hence, during this de-excitation process the electrons can emit the harmonic frequencies $\omega' = n\omega (n = 2, 3, \ldots)$. In accordance with the experimental set-up [14], we choose the vector of linear polarization of the laser field to be oriented perpendicular to the infinitely high potential wall. As it turns our, the laser-induced non-linear processes during electron reflection from the wall are strongly aligned along the vector of polarization of the field, whereas the free motion of the electrons along the other two space-directions is of minor importance [10]. Hence, we finally obtain a one-dimensional-model problem.

2. The model of harmonic production

With reference to the above discussion, we investigate a very elementary process of harmonic production, much simpler than the one considered by Sacks and Szöke [16]. An impenetrable potential wall is located in the (x, y)-plane such that $V(z) = 0$ for $z \leqslant 0$ and $V(z) = \infty$ for $z > 0$. Electrons impinge from the left in the e_z-direction and are reflected by the wall at $z = 0$. During this process the electrons are embedded in a powerful laser field, represented by a plane wave of frequency ω and polarization e_z. The wave is taken in the dipole approximation and the process is described non-relativistically. This essentially one-dimensional laser-modified scattering process has been investigated by Berson and Bondars [17] many years ago. We consider, in addition, the radiation spontaneously emitted during the scattering. If the total-, initial- and final-electron states are the same, as discussed in the introduction, the emitted radiation will correspond to harmonic production which, as will be illustrated below, shows plateau formation. Describing the radiation field by $\boldsymbol{A}(t) = (cF/\omega)\boldsymbol{e}_z \cos\omega t$, the Schrödinger equation for an electron in this field has the Gordon–Volkov solution [18, 19]

$$\psi_p = \exp\left[-(i/\hbar)(Et - p(z - \alpha\sin\omega t))\right] \tag{1}$$

where

$$\alpha = \mu c/\omega \qquad \mu = eF/mc\omega \tag{2}$$

with μ being the intensity parameter. The on-shell relation for the momentum p reads

$$p = [2m(E - \sigma)]^{1/2} \qquad \sigma = mc^2\mu^2/4 \tag{3}$$

where σ is the laser-induced AC-Stark shift. Apart from σ, the A^2 part of the interaction is neglected, since it does not contribute.

To fulfil the boundary conditions

$$\psi(z = 0, t) = 0 \tag{4}$$

at the wall, the exact total solution $\psi(z, t)$ of the scattering problem has to be a linear superposition of ingoing and reflected waves of the form (1). Hence, with reference to Berson and Bondars [17] and to our earlier work [20], we make the Floquet-type ansatz

$$\psi(z, t) = \psi_{p_0}^{(+)} - \sum_{n=-\infty}^{+\infty} R_n \psi_{p_n}^{(-)} \tag{5}$$

with

$$\psi_{p_0}^{(+)} = \exp\left[-(\mathrm{i}/\hbar)(E_0 t - p_0 z)\right]\exp\left[-\mathrm{i}a_0 \sin \omega t\right] \tag{6}$$

$$\psi_{p_n}^{(-)} = \exp\left\{-(\mathrm{i}/\hbar)\left[(E_0 + n\hbar\omega)t + p_n z\right]\right\}\exp\left[\mathrm{i}a_n \sin \omega t\right] \tag{7}$$

where

$$p_n = \left[2m(E_0 + n\hbar\omega - \sigma)\right]^{1/2} \qquad a_n = p_n\alpha/\hbar \tag{8}$$

$\psi_{p_0}^{(+)}$ represents an incoming electron of energy E_0 and momentum $\mathbf{p}_0 = p_0\mathbf{e}_z$, while $\psi_{p_n}^{(-)}$ are the components of the reflected electron wave of energies $E_0 + n\hbar\omega$ and momenta $\mathbf{p}_n = -p_n\mathbf{e}_z$. For $E_0 + n\hbar\omega - \sigma < 0$; p_n is purely imaginary ($\mathrm{i}\pi_n$) and these components belong to closed channels, which nevertheless contribute appreciably. Hence, there will be a smallest integer number $n_0 \gtrless 0$ for which p_{n_0} is real, while p_{n_0-1} is imaginary ($\mathrm{i}\pi_{n_0-1}$). Hence, if $n < n_0$ we obtain evanescent waves.

Applying to (5) the matching condition (4) and expanding (6) and (7) in terms of ordinary Bessel functions J_n, we find for the reflection coefficients R_n the matrix relation

$$J_n(a_n + a_0) = \sum_{k=-\infty}^{+\infty} J_{n-k}(a_n - a_k)R_k \tag{9}$$

which, in general, has complex matrix elements and has to be solved numerically for the reflection coefficients R_n. Moreover, this matrix equation represents a complicated, multi-channel interference problem. Therefore, certain sharp irregularities in the electron reflection probability currents presented below in figure 1 for higher laser field intensities are hard to explain by some analytic considerations. In the low frequency and low intensity approximation only, we get $R_n \simeq J_n(a_n + a_0)$, which is the first Born amplitude. Having evaluated the reflection coefficients R_n, the renormalized components of the probability current associated with the different n-photon back-scattering processes are given by $j(n) = p_n(|R_n|^2)/p_0$ and satisfy the sum rule

$$\sum_{n=n_0}^{+\infty} j(n) = 1. \tag{10}$$

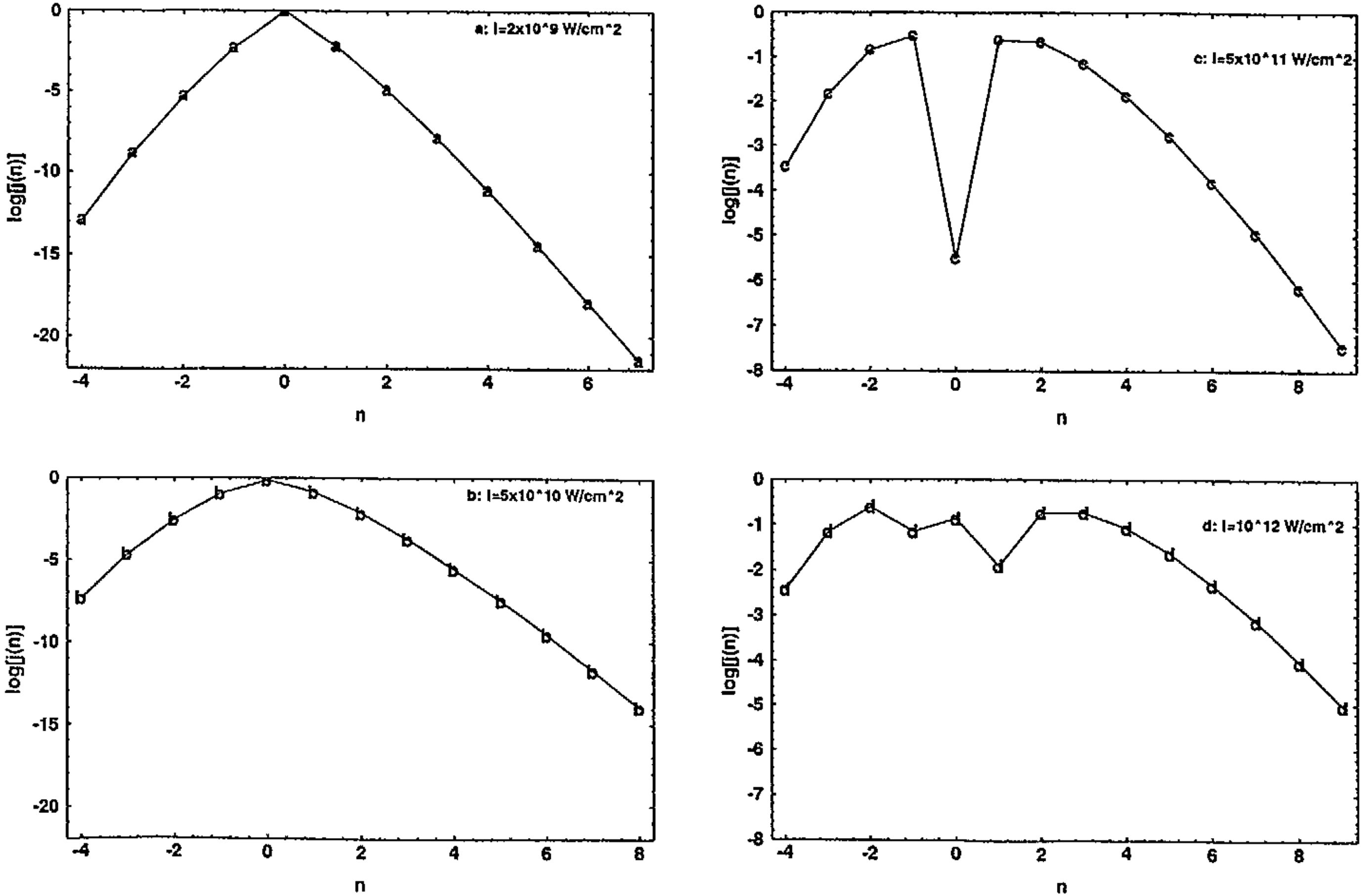

Figure 1. In this figure we show $\log_{10}[j(n)]$ as a function of n for the laser intensities (a): $I = 2 \times 10^9$ W cm^{-2}, (b): $I = 5 \times 10^{10}$ W cm^{-2}, (c): $I = 5 \times 10^{11}$ W cm^{-2} and (d): $I = 10^{12}$ W cm^{-2}. Observe in the case (c) the drastic decrease of $j(0)$.

Turning next to the emission of harmonic photons of polarization ϵ' and wavevector $\boldsymbol{k}'$ produced during the scattering in the laser field, the responsible interaction term in dipole approximation (in the $\boldsymbol{E}\cdot\boldsymbol{x}$-gauge) is given by

$$-\,\mathrm{i}e\big(2\pi\hbar\omega'/L^3\big)(\epsilon'\cdot\boldsymbol{x})\exp(\mathrm{i}\omega' t) \tag{11}$$

where $V = L^3$ is the normalization volume. Hence, for calculating the production of higher harmonics we have to evaluate the expectation value $\langle\psi|\epsilon\cdot\boldsymbol{x}|\psi\rangle = \epsilon'_z\langle\psi|z|\psi\rangle$. Fortunately, by using the total wavefunction ψ from (5)–(7) a necessary gauge factor (relating the $\boldsymbol{p}\cdot\boldsymbol{A}$-gauge with the $\boldsymbol{E}\cdot\boldsymbol{x}$-gauge) drops out so that we find by means of the Fourier expansion of the expectation value

$$\langle\psi|z|\psi\rangle = \langle z\rangle(t) = \sum_{n=-\infty}^{+\infty} z_n \exp(-\mathrm{i}n\omega t) \tag{12}$$

the following representation for z_n with $n \geqslant 2$

$$\begin{aligned} z_n = \big(\hbar^2/L\big)\Bigg\{&-2\mathrm{Re}\Bigg[\sum_{k=-\infty}^{+\infty} J_{n-k}\big(-a_k + a_0\big)R_k/\big(p_k + p_0\big)^2\Bigg] \\ &+ \sum_{k\neq l=-\infty}^{+\infty} J_{n+k-l}\big(a_k^* - a_l\big)R_k^* R_l/\big(p_k^* + p_0\big)^2 \\ &+ \sum_{k=-\infty}^{n_0-1} J_n\big(-2a_k\big)\big(|R_k|^2\big)/4p_k^2\Bigg\}. \end{aligned} \tag{13}$$

In deriving (13), we used the relation $\int_{-\infty}^{0} \mathrm{d}z \exp(\mathrm{i}kz) = \pi\delta(k) - \mathrm{i}P(1/k)$ and it turned out that the terms proportional to the δ-function do not contribute to the harmonic generation. We should observe that of the total real expectation value in (12) only those Fourier components z_n are essential, which yield generation of harmonics. These are found by combining (11) and (12) and integrating over t, in which case a term $\delta(\omega' - n\omega)$ appears, *being equal to zero for* $n \leqslant 0$.

Introducing the dimensionless quantities $\bar{z}_n = Lk_0^2 z_n$, with $k_0 = p_0/\hbar$ being the wavenumber of the incoming electron, we obtain from (11) and (13) for the transition probability per unit time and unit solid angle for the nth harmonic production

$$\partial^2 P_n/\partial\Omega\partial t = \big(\epsilon'_z\big)^2\big(e^2/\hbar c\big)\big(\omega^3/2\pi c^2\big)\big(Lk_0^2\big)^{-2} n^3|\bar{z}_n|^2 \tag{14}$$

where the n-dependence of the production rates for emission by a single electron is governed by the quantity [1]

$$w(n) = n^3|\bar{z}_n|^2 \qquad n = 2, 3, \ldots. \tag{15}$$

We should observe that due to the lack of symmetry even and odd harmonics are produced.

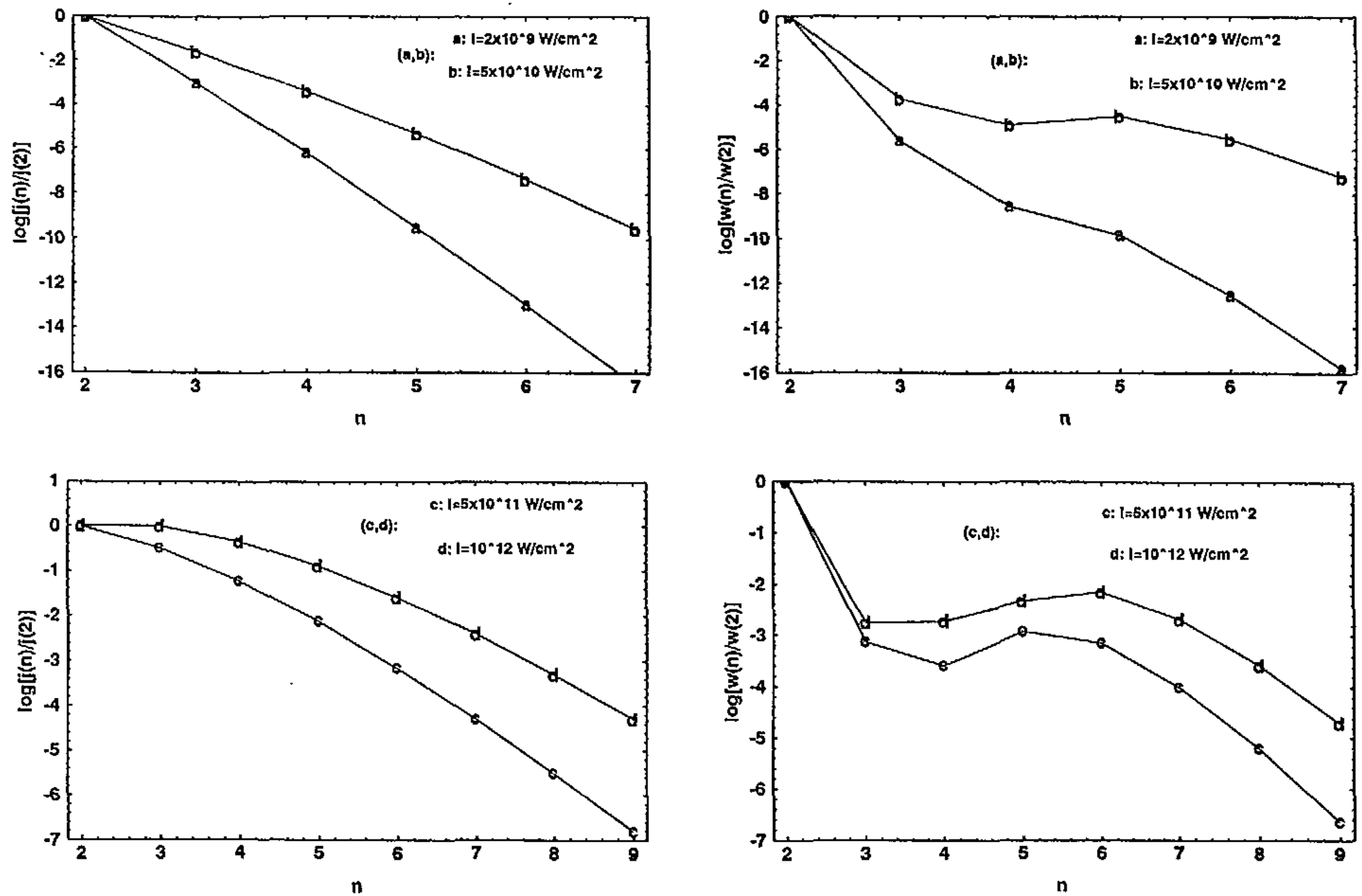

Figure 2. Here we show for the same intensities as in figure 1 on the left hand side $\log_{10}[j(n)/j(2)]$ and on the right-hand side $\log_{10}[w(n)/w(2)]$. Starting in figure 2(b), observe the plateau formation of the harmonic rates.

3. Numerical example

In the following figures we display on a logarithmic scale the probability currents $j(n)$ of electron back scattering with the stimulated emission or absorption of n photons for $n \geqslant n_0$. We also present the $\log_{10}$ of $j(n)/j(2)$, to be compared with the corresponding logarithm of the relative harmonic production rates $w(n)/w(2)$ as a function of the harmonic order n. As initial electron energy we choose the Fermi-energy of gold $E = E_F = 5.51$ eV and we assume irradiation with an Nd:YAG laser with $\hbar\omega = 1.17$ eV in accordance with the experiment of Farkas *et al* [14]. The laser field intensities considered are: (a) $I = 2 \times 10^9$ W cm^{-2}, (b) $I = 5 \times 10^{10}$ W cm^{-2}, (c) $I = 5 \times 10^{11}$ W cm^{-2} and (d) $I = 10^{12}$ W cm^{-2} which is within the limits of intensities used by Farkas and co-workers in their more recent experiments [15]. In all four cases the lowest open channel is $n_0 = 4$. Therefore, electron waves with $n \geqslant -4$ and thus real momenta p_n can propagate, whereas those with $n < -4$ have imaginary momenta $p_n = \mathrm{i}\pi_n$ and correspond to evanescent waves. In figure 1 we show $\log_{10}[j(n)]$ as a function of n for the above laser intensities (a), (b), (c) and (d) and in figure 2 we present on the left-hand side for the same intensities $\log_{10}[j(n)/j(2)]$ with $n \geqslant 2$, which data should be compared with the corresponding values of $\log_{10}[w(n)/w(2)]$ on the right-hand side, where in the cases (a): $w(2) = 1.71 \times 10^{-7}$, (b) $w(2) = 1.11 \times 10^{-4}$, (c) $w(2) = 1.11 \times 10^{-2}$ and (d) $w(2) = 4.67 \times 10^{-2}$. For the lowest laser intensities, the data in all these figures roughly follow the power law $I^n (n \geqslant 2)$, but this is evidently no more the case for the higher intensities, indicating the necessity of exactly evaluating the reflection coefficients R_n by a numerical inversion of (9) and by then using these data in (13). The most prominent feature of the calculated data, however, is the formation of a plateau of the harmonic production rates, plotted as a function of the harmonic order n, displayed on the right-hand side of figure 2. These findings are in contrast to what has been obtained in the work of Sacks and Szöke [16], who considered harmonic production during scattering of electrons by a one-dimensional, piece-wise constant potential, where no plateaux showed up in the harmonic rates.

We conclude that the confined motion of electrons in a deep potential well has some influence on the plateau formation of the rates of the harmonics produced during the reflection of these electrons from one of the walls of the well in a laser field. In our artificial model metal, the Fermi-energy E_F takes the role of the ground-state energy of the system to which the electrons fall back after their excitation by the laser field to emit the harmonic frequencies $\omega' = n\omega$ $(n = 2, 3, \ldots)$. For higher laser field intensities, the distribution of the harmonic rates, shown on the right-hand side of figure 2, has a cut-off which roughly increases linearly with the laser intensity I. For an intensity of about 2×10^9 W cm^{-2}, when the plateau starts to develop, this cut-off is nearly at $n = 5$. This corresponds to an emitted photon energy $\hbar\omega' = 5.85$ eV being nearly equal to the Fermi-energy of gold $E_F = 5.51$ eV and also close to the work function of gold $W = 4.6792$ eV. This is in agreement with the cut-off frequency observed by Farkas and co-workers [14] at the above laser field intensity and appears to be independent of the fact that our model metal cannot be ionized. For real metals, of course, the concomitant laser-induced multiphoton photo effect has been observed by several research groups [21–23], but no satisfactory explanation has yet been found for the rather high energy electrons of some 300–500 eV emitted during this process at moderate laser field intensities [22, 24]. For atoms as well as for real metals, it is quite clear from simple unitarity considerations of the scattering operator S that multiphonon ionization and harmonic generation should be competing processes [25].

4. Concluding remarks

In the foregoing sections we considered the generation of harmonics irradiating a metal surface with laser light. Our metal has the rather artificial property of having an infinitely large work function W. Describing the metal by Sommerfeld's free electron model, this means that the free electron gas, obeying the Fermi–Dirac statistics, is enclosed in a potential well of infinitely high walls. Since the laser field can only penetrate into the metal within a thin layer, the interaction of the electrons with the field can only take place during their reflection from one of the walls of the well, fulfilling the proper boundary condition at the wall. The dipole moment of the corresponding total wavefunction is then responsible for the generation of harmonics. Moreover, on account of the properties of the Fermi–Dirac probability distribution, only those electrons will be involved in the harmonic generation process, whose energy is close to the Fermi-energy E_F (to simplify the calculation we took $E_F(T = 0)$. E_F then acts as the ground-state energy of the system to which the electrons, excited by the laser field, return and emit the harmonic frequencies $\omega' = n\omega$ ($n = 2, 3, \ldots$). In section 2 we found that for this model of a metal the harmonic rates, plotted as a function of the harmonic order n, show at higher laser intensities the formation of a plateau, as can be seen on the right-hand side of figure 2. On the other hand, for real metals no plateau formation has been observed experimentally [14, 15]. In these experiments, however, rather high laser frequencies have been used such that $n\hbar\omega < W$, where n is a small positive integer. Contrary to these findings, we expect on the basis of our considerations that for much lower laser frequencies with $\hbar\omega \ll W$, the formation of a plateau will appear in the rates of harmonic generation as obtained from laser irradiation of a metal surface. So we essentially predict that the plateau formation for a metal depends on the ratio $\hbar\omega/W$. Of course, in all such experiments the laser pulses would have to be sufficiently short in order to prevent the formation of a surface plasma.

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