

High Order Harmonic Generation from a One-Dimensional Solid

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In the presence of the intense laser field, the high-order harmonic generation spectrum in the one-dimensional crystals has been presented for the laser wavelength $\lambda = 780$ nm. The calculated emission strength for three types of crystals (insulator, metal, and semiconductor) are compared. The semiconductor medium and/or metallic crystal are found to be more efficient than insulator.

1. Introduction:

High-order harmonic generation is a promising technique for obtaining new sources of coherent radiation because not only does it provide a probe for investigating the interaction mechanism of the solid system with an intense laser field, but it also has the potential to become the future coherent XUV source.

Sacks and Szoke [1] have studied electron scattering from a piecewise-constant potential in an intense electromagnetic field, and they reported the generation of harmonics and evaluated the rates of high harmonic production for an arbitrary order of perturbation theory. Also, the multi-harmonic generation and multi-photon electron emission at a metal surface have been studied by Mishra and Gresten [2,3] on the basis of free electron theory of Sommerfeld.

Theoretical study of the high harmonic generation in crystalline solid by non-perturbative method based on the numerical computation of solutions of the Schrodinger equation is given by Plaja *et al.* [4]. Also, the mechanisms to enhance harmonic generation without excessive excitation of electrons to the conduction band are given. Huller *et al.* [5] investigated high-order harmonic generation from a thin solid layer illuminated by a subpicosecond laser pulse. Recently, Faisal el al. [6,7] have shown by non-perturbative (Floquet-Bloch) simulations that it is possibly in principle, to generate very high harmonics via

the mechanism of interband resonance, using only moderately intense laser fields that interact with transmission Bloch electrons in a thin crystal.

In this work, the high-order harmonic generation spectrum for three types of crystals (insulator, metal, and semiconductor) will be calculated.

2. Theory:

2.1. Modified Band Structure:

Quantum mechanically and in the presence of intense monochromatic laser field, a one-dimensional solid treated as a periodic array of single quantum wells (Kronig-Penney model [8] with the Hamiltonian (in atomic units $e=m=\hbar=1$)

$$H(t) = H_0 + 1/c(A(t) \cdot p) + (1/2c^2)A^2(t). \quad (1)$$

with H_0 is the unperturbed Hamiltonian given by

$$H_0 = p^2/2 + V(x), \quad (2)$$

where $v(x)$ is the electrostatic potential and will be expressed by the well-known Kronig-Penney periodic delta potential

$$v(x) = \beta \sum_{j=0}^{j=\infty} \delta(x - ja) \quad (3)$$

of strength $\beta = P/2a$ with lattice constant a . The laser field is assumed to be linearly polarized along the crystal axis in the x -direction and is given by the vector potential

$$A(t) = A_0 \cos(\omega t + \delta), \quad (4)$$

where A_0 is related to the peak strength of the field E_0 by $E_0 = \omega A_0/c$; ω is the frequency of the monochromatic light and δ is an arbitrary phase. This model has been first studied approximately by Tsoar and Gersten [9] who obtained the dispersion relation of the system in terms of a double-infinite matrix and discussed the modification of the band structure with approximate calculations. It was shown [10] how to obtain an exact analytic solution of the dispersion relation in terms of a single-infinite matrix. The problem can be solved to obtain the wave function $\Psi_{\mathbf{k}}(x, t)$ as follows:

$$\Psi_{\nu k}(x, t) = -i\beta e^{-i(E_\nu(k)+\delta_E)t} \sum_{N=-\infty}^{j=\infty} \frac{1}{kN} \{ \cos ka - \cos k_\nu Na \} C_N(E_\nu(k))$$

$$N = - \sum_{N=-\infty}^{\infty} J_{l-N}(\alpha_0 k_\nu e^{i(\omega t + \delta)}) \sum_{j=-\infty}^{\infty} e^{ikN + ikja|x-ja|} \quad (5)$$

where

$$k_{\nu N} = \sqrt{2(E_\nu(k) - N(\omega))} \quad (6)$$

and

$$(7)$$

E(k) is the eigenvalues of the problem, $b = \delta_N / 2\omega = A_0 / 4c^2$ and the constant $\alpha_0 = \frac{A_0}{\omega c} = \frac{E_0}{\omega^2}$, is the classical radius of vibration of the electron in the field, the so called "quiver radius". The generalized Bessel function J (a-b) is used as in the following

$$J_p(a-b) = \sum_{m=-\infty}^{\infty} J_{p+m}(\alpha) j_m(b) \quad (8)$$

In the presence of the field $E_0 \neq 0$, the quasienergy E(k) fulfills the exact dispersion relation, $\det[(\cos ka - \cos kNa)\delta_{NN}]$.

$$- \frac{\beta}{K_N} J_{N-N}(\alpha_0(k_N - k_{N'})) \sin k_N a = 0 \quad (9)$$

The eigenvalues E(k) of equation (9) appear in the parameters k_N in equation (6) as a function of k.

2.2. High-order harmonic generation:

The harmonic generation is the rate of generating photons at a frequency equal to a harmonic of incident laser which is related to the expectation value of the probability current density integrated over the elementary cell, and for the 1-D crystal it can be written down as[11,12]

$$\frac{ds}{d\Omega} = \frac{\alpha^3}{3\pi} \left[\sum_{\nu} \sum_{|k| \leq k_F} \int_{-\frac{T_0}{2}}^{\frac{T_0}{2}} dt e^{i\Omega t} \left[\frac{\partial}{\partial t} N_c \int_0^a dx j_{\nu k}(x, t) \right] \right]^2 \quad (10)$$

in which N_c the effective length of the crystal, N_c is the number of unit cell in the crystal, $\alpha=1/137$ is the fine structure constant, T is the time duration of the field pulse and the probability current density is written as

$$j_{vk}(x,t) = \frac{1}{2} [\Psi_{vk}^*(x,t) (\frac{1}{i} \frac{\partial}{\partial x} - \frac{1}{c} A(t) \Psi_{vk}(x,t)) + \Psi_{vk}(x,t) (\frac{1}{i} \frac{\partial}{\partial x} + \frac{1}{c} A(t) \Psi_{vk}^*(x,t))] \quad (11)$$

The summation extends over quasimomenta of the occupied bands, and over such quasimomenta of the partially occupied band that are smaller than the Fermi momentum k_F . By substituting $\Psi_{vk}(x,t)$ as in [5] and using in the dipole approximation as in [4] and Fourier analyze the probability current density integrated over an elementary cell to get

$$\int_0^a \Psi_{vk}(x,t) dx = \sum_{N'} e^{iN'(\omega t + \delta)} \zeta_{vN'}(k) \quad (12)$$

with the Fourier components

$$\zeta_{vN'}(k) = \frac{1}{2} \beta^2 N_{vk}^{-1} \sum_{N,M=-\infty}^{\infty} \frac{C_N(E_v(k)) C_M^*(E_v(k))}{k_{vN} k_{vM}^*} (\cos ka - \cos k_{vN} a) (\cos ka - \cos k_{vM}^* a) \sum_{j,j'=-\infty}^{\infty} e^{ik(j-j')a} I_{NMjj'}[\Phi] \quad (13)$$

where

$$\Phi = (\varepsilon k N + k_{vM}^* M) J_{M-N-N'}(\alpha_0(k_{vN} - k_{vM}^*)) - \alpha_0 \omega (J_{M-N-N'+1}(\alpha_0(k_{vN} - k_{vM}^*)) + J_{M-N-N'-1}(\alpha_0(k_{vN} - k_{vM}^*)))$$

and N_{vk} is the normalization constant

$$N_{vk} = \beta^2 \sum_{N,M=-\infty}^{\infty} \frac{C_N(E_v(k)) C_M^*(E_v(k))}{K_{vN} K_{vM}^*} (\cos ka - \cos k_{vN} a) (\cos ka - \cos k_{vM}^* a) J_{M-N}(\alpha_0(k_{vN} - k_{vM}^*))$$

$$, \quad \sum_{j,j'=-\infty}^{\infty} e^{ik(j-j')a} I_{Nmjj'} \quad (14)$$

and

$$I_{NMjj'} = \frac{1}{i(\varepsilon_1 k_{vN} - \varepsilon_2 k_{vM}^*)} [e^{i\varepsilon_1 k_{vN} ja - i\varepsilon_2 k_{vM}^* (1-j)a} - e^{i\varepsilon_1 k_{vN} (1-j)a - i\varepsilon_2 k_{vM}^* j'a}] \quad (15)$$

where

$$\varepsilon_1 = +1 \quad \text{for } j \leq 0 \quad \text{and} \quad \varepsilon_1 = -1 \quad \text{for } j > 0$$

and

$$\varepsilon_2 = +1 \quad \text{for } j' \leq 0 \quad \text{and} \quad \varepsilon_2 = -1 \quad \text{for } j' > 0$$

in the limit of large T_0 , the rate of emission of high harmonic radiation $dW/d\Omega$, at the frequency Ω takes the form,

$$\frac{dW}{d\Omega} = \frac{1}{T_0} \frac{ds}{d\Omega} = \frac{2}{3} \alpha^3 N_c^2 \sum \delta(\Omega - N\omega) S_{N'}(k_F) \quad (16)$$

or integrating over the frequency interval $d\Omega$,

$$W = \frac{2}{3} \alpha^3 N_c^2 \sum S_{N'}(k_F) \quad (17)$$

where

$$S_{N'}(k_F) = \left| \sum_{v'} \sum_{|q| \leq q_f} D_{vN'}(q) \right|^2, \quad (18)$$

$D_{vN'}(q)$ is the current distribution function,

$$D_{vN'}(q) = N\omega \varepsilon_{vN'}(k) \quad (19)$$

with $q = ka$ is the dimensionless quasi-momentum. The squared Fourier components $S_{N'}(k_F)$ of the current contain the essential information of the dynamics and the rest of the factors in (17) are mere constants.

3. Results and Discussions:

In our analysis, we assume the lattice constant $a=8.5a.u.$ and the mean strength of the potential $P = 7$. These parameters have been chosen such that for a one-and-half filled crystal, i.e. for the fully filled first band and the half-filled second band, the Fermi energy is equal to $5.53eV$ and the work function $W = 5.1 eV$, these are relevant parameters for a gold crystal[13] which was used in the experiment of Farkas et al [14].

The laser field is characterized by its frequency ω and its amplitude E_0 which determines the intensity $I = cE_0^2 / 8\pi$ (c is speed of light). In this investigation we assume the laser photons to have the wavelength $\lambda = 780nm$ ($\omega = 0.0584 a.u.$).

The high harmonic generation spectrum, $S_N(k_F)$, has been calculated for three types of crystals. These types of crystals are (a) insulator when only the first band is occupied and there is a wide gap between the valence band and the conduction band, (b) metal when the second band is half-filled by electrons, and (c) semiconductor when the second band is completely filled and there is a narrow gap between the valence and the conduction bands. The results obtained at different intensities of the laser field in the range between $I=3.51 \times 10^{10} W/cm^2$ and $I=1.755 \times 10^{12} W/cm^2$ are presented in Figs. (1) for an insulator, fig(2) for a metal and fig (3) for a semiconductor. From the inversion symmetry of the periodic structure, only odd harmonics N are considered here.

At relatively low intensity $I = 3.51 \times 10^{10} W/cm^2$ ($10^{-6} a.u.$), the strength of the emitted power spectrum drops linearly (in the logarithmic scale) with the order N which is consistent with the expectation based on perturbation theory, i.e., with the power law

$$S_N(k_F) = S_0 (I / I_{sat})^N. \quad (20)$$

where S_0 is a constant and I_{sat} is called the saturation intensity for high harmonic generation.

From eq (20) we have

$$\text{Log} S_N(k_F) \approx N \text{Log}(I / I_{sat}) + \text{log } S_0 \quad (21)$$

from the slope of the lines, it can be seen that $\text{log}(I/I_{sat}) = -2$. Thus, the saturation intensity (I_{sat}) in a solid, irrespective of the field-free conduction

properties, is of the order 10^{-4} a.u. (3.51×10^{12} W/cm²), i.e., much lower than for the most noble gas targets, for which, such intensities are still perturbative in nature.

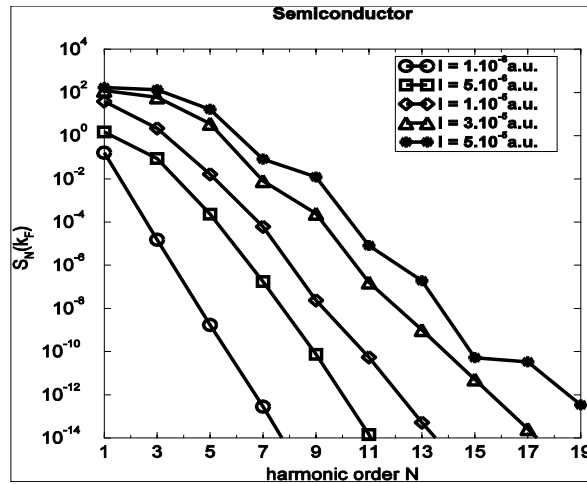


Fig. (1): high harmonic generation power spectrum for a 1-D crystal at a laser field wavelength $\lambda = 780$ nm and with different intensities. Results are shown for Fermi momentum corresponding to an insulator.

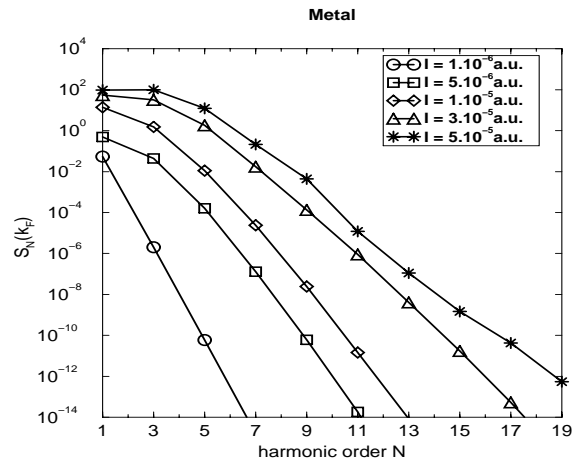


Fig. (2): high harmonic generation power spectrum for a 1-D crystal at a laser field wavelength $\lambda = 780$ nm and with different intensities. Results are shown for Fermi momentum corresponding to a metal.

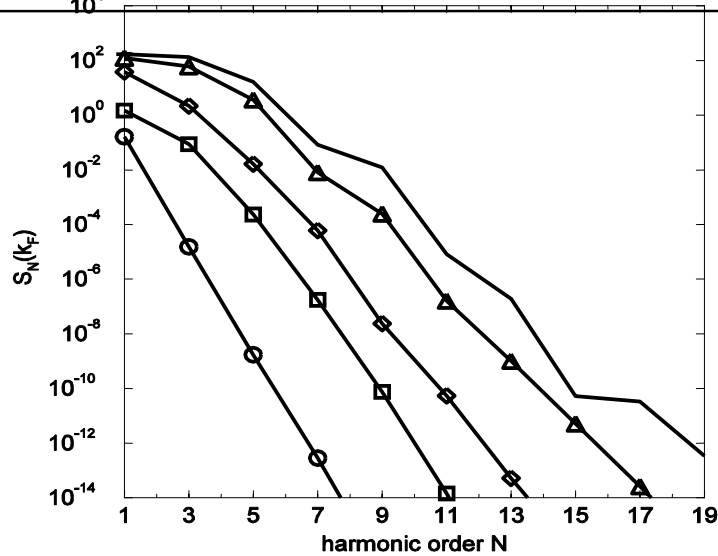


Fig. (3): high harmonic generation power spectrum for a 1-D crystal at a laser field wavelength $\lambda = 780$ nm and with different intensities. Results are shown for Fermi momentum corresponding to a semiconductor.

The non-perturbative effects are already seen (in reference [12] using a more generalized model) for different larger intensities $I = 1.755 \times 10^{11}$, 3.51×10^{11} , and $1.053 \times 10^{12} \text{ W/cm}^2$, where the third harmonic is comparable with the first one, and for a very large intensity $I = 1.755 \times 10^{12} \text{ W/cm}^2$ (5×10^{-5} a.u.) the third harmonic is stronger than the first one. At this high intensity 5×10^{-5} a.u., it is also seen that for semiconductors the ninth harmonic is comparable with the seventh one and the 17th harmonic is stronger than the 15th one, whereas for metals or insulators the fifth harmonic is comparable with the third. It is seen that increasing the intensity significantly increases the emission strength for all harmonics and for types of crystals, insulators (1), metals (2), and semiconductors (3). It is noticed that as the intensity of the laser field is lower than the 10^{10} the used model gives different results from that in reference [12]. At a higher intensity the decrease of the emission strength with increasing order N of the harmonics is significantly slower than at lower intensities. The higher intensity results also show a non-perturbative behavior in that two neighboring harmonics sometimes appear with comparable strength, an effect which does not occur at the lower intensities. Finally, it can be seen that for the lower order harmonics a semiconductor and/or a metallic crystal is somewhat more efficient than the insulator.

4. Conclusion:

In conclusion, the power spectrum of high-order harmonic generation is calculated for three types of crystals (insulator, metal, and semiconductor). It is shown that insulators, metals and semiconductors are much efficient than atomic gas in generating high harmonics at moderate intensities. For wavelength $\lambda = 780$ nm at intensity $I = 1.755 \times 10^{12}$ W/cm², a semiconductor and/or metallic crystal is comparatively more efficient in generating higher harmonics than insulators.

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