Generation and control of high harmonics by laser interaction with transmission electrons in a thin crystal

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We propose a mechanism of generating very high harmonics at moderate laser intensities by transmission of electrons in a thin crystal. Results of nonperturbative Floquet-Bloch calculations show the possibility of controlling the height and range of high-harmonic spectra through the mechanism of interband resonances at suitable energies of the incident electrons and/or intensity of the laser field. [S1050-2947(96)50809-7]

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Discovery of high-harmonic generation [1,2] by the interaction of intense laser light with gas atoms has stimulated much interest in the investigation of analogous processes in crystalline media. In fact, the first observations of high-harmonic emission in crystals, with an efficiency higher than in gas media, have recently been reported [3]. Thus, harmonics up to the fifth order have been observed, by Farkas and co-workers, from gold targets, with picosecond neodymium-doped yttrium aluminum garnet (Nd:YAG) lasers at intensities in the region of 10^9 W/cm^2; and up to the 15th order, by von der Linde and co-workers, from an aluminum target, with a femtosecond Ti:sapphire laser at intensities of the order of 10^{17} W/cm^2.

In this Rapid Communication we suggest, on the basis of a nonperturbative theoretical investigation, a mechanism of generation, and possibly control, of very high harmonics in crystals using only moderately strong laser fields. To this end we consider the interaction of a laser field with a beam of electrons in transmission through a pure thin crystal or film. It is found that the high-harmonic spectra originating from the transmitting Bloch electrons of appropriate energy and momenta can give rise to a remarkable enhancement of higher harmonics through the presence of interband multiphoton resonances. These resonances may be induced either by increasing the intensity (at a fixed frequency) or by suitably tuning the incident energy of the electrons.

Before proceeding further, let us qualitatively estimate some of the macroscopic parameters involved for a potential realization of the process in the laboratory. The skin depth of electromagnetic radiation in a metal crystal is of the order of 170 Å. Thus, we assume the width of the thin crystal (or film) to be, say, 100 Å, so that even a grazing incidence of the light beam may permit effective interaction with the electrons over the entire width of the crystal. We also assume a highly pure crystal, so that at electron energies of the order of, say, 20 eV, the absorption length has to be between 50 and 100 Å, and so that a significant part of the incident electron beam may not be scattered away incoherently before crossing the crystal width. Alternatively, instead of using an electron beam from outside, the Bloch electrons may be prepared initially in the appropriate empty bands by pumping the crystal with a high-frequency light source and/or with the intense laser field itself [4]. To estimate the laser pulse duration and the peak intensity that may not be exceeded in such experiments, we note that for an atomic layer of moderate Z materials of 100 Å the lattice disintegration time, in the field of a Nd:YAG laser of 10^{15} W/cm^2, has been estimated to be several hundred femtoseconds [5] and the electron-phonon relaxation time [6] to be a few hundred femtoseconds. One may thus allow a rather high-intensity (10^{15} W/cm^2) Nd:YAG laser pulse of the order of 100 fs, without unduly damaging the crystal lattice. But perhaps a more severe restriction on the intensity of the field is imposed by the characteristic intensity for ionization breakdown of the crystal. There are at present no definitive estimates of the same available. We may, however, get an order of magnitude estimate by requiring the onset of the ATI (above threshold ionization) process, for this purpose. The ATI threshold occurs generally for $U_p > \hbar\omega$, where U_p is the ponderomotive energy. This gives for a Nd:YAG laser an intensity greater than 10^{13} W/cm^2. We shall therefore restrict ourselves to intensities under this value for the results of the simulations to be discussed below.

Theoretical investigations of high-harmonic generation in gases have been made extensively in the past (e.g., [7]) whereas corresponding studies in crystalline media have been initiated only recently [8,9]. For the present purpose we have applied a nonperturbative Floquet-Bloch analysis to solve the associated Schrödinger equation of the system. The method of solution has been discussed elsewhere [10,11,9], and hence need not be repeated here. However, we point out briefly the basic assumptions of the model used. It is assumed that the motion of the transmission electrons is determined by a periodic sequence of square wells along the crystal axis aligned parallel to the polarization vector of the laser. For the purpose of the simulations, the depth, the width and the period length of the crystal potential are chosen to be -7.75 eV, 1.14 Å, and 4.8 Å, respectively. This gives, for example, a Fermi energy of 5.53 eV, which compares well with that of gold.

The band structure of the model system calculated in the absence of the laser field in Fig. 1 that shows the lowest eight bands of the system, as a function of the dimensionless quasimomentum $q = kl$ (k is the Bloch vector and l is the

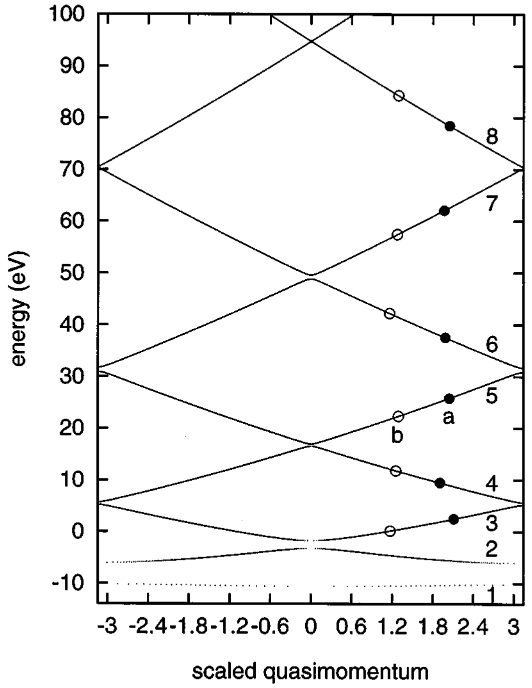

FIG. 1. Band structure of the model crystal in the absence of the field showing the first 8 bands. The points *a* and *b* refer to two selected electron energies. The filled and empty circles indicate the positions of interband resonance energies.

lattice period), is restricted to the first Brillouin zone: $-\pi<q<\pi$. From this figure we can see that an electron with an energy on the order of 20 eV, and its momentum component along the crystal axis correponding to a Bloch

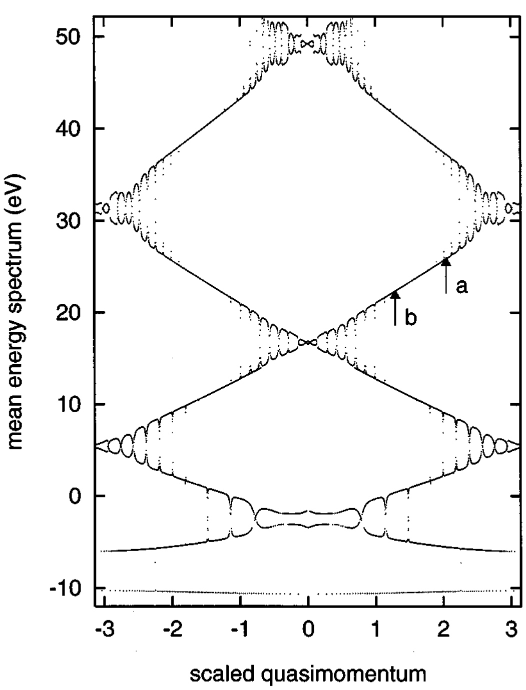

FIG. 2. Mean energy spectrum showing the lowest 6 bands in the presence of the field, $\omega=1.169$ eV and $I=3.51\times10^{11}$ W/cm^2. The arrows *a* and *b* indicate the two selected energies of the transmission electrons.

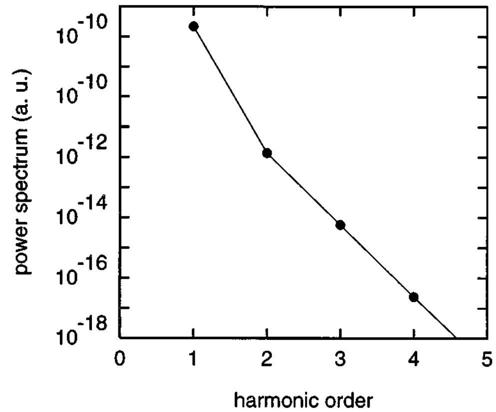

FIG. 3. Harmonic generation spectrum at a perturbative intensity $I=3.5\times10^{9}$ W/cm^2, $\omega=1.169$ eV, and incident electron energy $E_i=25.8$ eV.

momentum, would propagate in the *fifth* band as a Bloch wave [12].

In the presence of the laser field each Bloch state evolves into a Floquet-Bloch state of the form (in a.u.) [10,11]

$$\psi_{\beta k}(x,t)=e^{-iE_{\beta}(k)t}e^{ikx}\phi_{\beta k}(x,t), \tag{1}$$

where $\beta=1,2,3,\ldots$, is the band index, $E_{\beta}(k)$ is the Floquet-Bloch eigenenergy, and $\phi_{\beta k}(x,t)$ is a periodic function of both x and t with periods $2\pi/l$ and $2\pi/\omega$, respectively. The resulting modification of the band structure by the field can be seen in the mean energy spectrum obtained from the expectation value of the energy operator in the Floquet-Bloch state, Eq. (1).

In Fig. 2 we show the mean energy spectrum for a Nd:YAG laser of frequency $\omega=1.169$ eV and intensity $I=10^{-5}$ a.u. (3.51×10^{11} W/cm^2). The most interesting features seen in Fig. 2 are the rather sharp modifications of the band structure near a multiphoton resonance between a pair of bands (two of which are marked as *a* and *b* in the figure).

The high-harmonic emission spectra of interest are obtained from the Fourier transform of the expectation value of

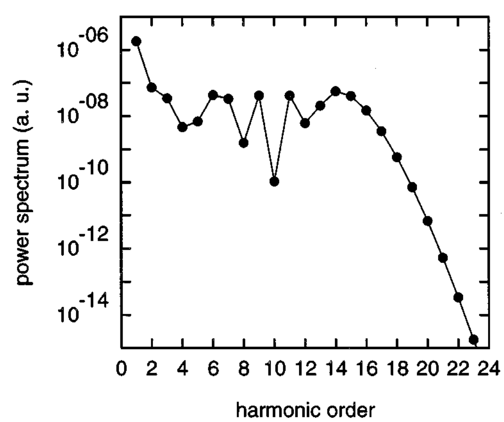

FIG. 4. High-harmonic spectrum at a nonperturbative intensity $I=3.5\times10^{11}$ W/cm^2, $\omega=1.169$ eV, and incident electron energy $E_i=$ 25.8 eV, showing the appearance of a ''plateau.''

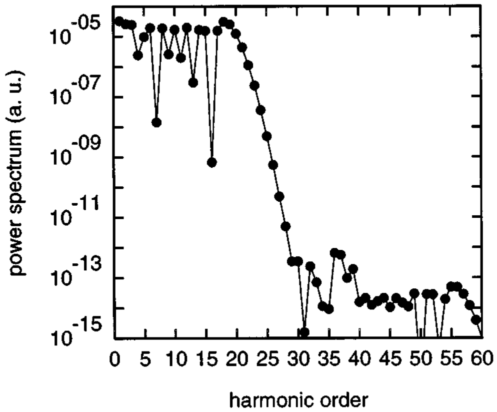

FIG. 5. The same as in Fig. 4, except $I=1.1\times10^{12}$ W/cm^2. Note the formation of the second plateau and the greatly enhanced height of the first plateau.

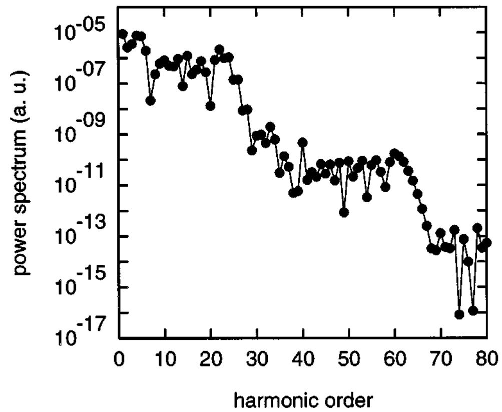

FIG. 7. The same as in Fig. 5 except $E_i=22.3$ eV.

the current density calculated from the Floquet-Bloch states defined above. The expression for the rate of emission at the frequency Ω then takes the form [9]

$$\frac{dW}{d\Omega}=\frac{2}{3}\alpha^3N_c^2\sum_{N\in\mathcal{Z}}\delta(\Omega-N\omega)|D_{\beta N}(q)|^2, \qquad (2)$$

where $D_{\beta N}(q)$ is the Nth Fourier component of the current, N_c is the effective number of elementary cells in the laser focus, and α is the fine-structure constant. In the following sequence of figures we present the results of simulation of the high-harmonic generation spectra, W, in terms of the squared Fourier components of the current, $|D_{\beta N}(q)|^2$, that contain the essential information of the dynamics. Note that the rest of the factors in Eq. (2) are mere constants.

Figure 3 shows the calculated spectrum at the intensity $I=10^{-7}$ a.u. (3.51×10^9 W/cm^2). In this case the state of the incident electron, with energy $E_i=25.8$ eV, corresponds to the state marked by a in the band structure of Figs. 1 and 2. Note that both odd and even harmonics are generated in the present system, as one expects from the lack of inversion symmetry of the transmission geometry. It can be seen from Fig. 3 that at $I=10^{-7}$ a.u., the harmonic strengths fall off linearly (in the logarithmic scale) with the order N, which is typical of the perturbative region of intensity. Dramatic changes in the behavior of the harmonic generation spectrum at nonperturbative intensities can be seen in Figs. 4–6. At an intensity 3.51×10^{11} W/cm^2, which is 100 times larger than the previous perturbative case, there appears a plateau (Fig. 4) in the spectrum, which extends up to the 14th harmonic. At a still higher intensity, $I=3\times10^{-5}$ a.u. ($\approx10^{12}$ W/cm^2), we observe (Fig. 5) an increase of the height of the plateau by more than two orders of magnitude compared to the previous case. More interestingly there also appears a *second* plateau, extending between the 32nd to the 56th harmonic. This new plateau is a consequence of resonant coupling of the incident 5th band with the 4th and the 3rd empty bands of the crystal (cf. Fig. 1). The multiphoton resonant coupling creates a significantly large amplitude for the corresponding Fourier component in the Floquet-Bloch state. This in turn leads to large amplitudes in the current density oscillating at various difference frequencies associated with the product of the resonant and the nonresonant Fourier amplitudes. These oscillations are then radiated into the higher-harmonic modes.

For the highest intensity considered in this investigation, $I=6\times10^{-5}$ a.u. ($\approx2.1\times10^{12}$ W/cm^2), the height of the second plateau is seen to rise (Fig. 6) by some three orders of

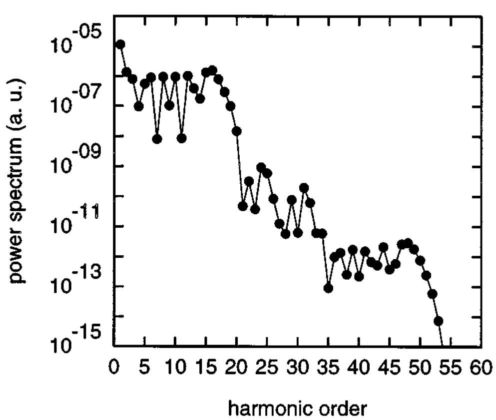

FIG. 6. The same as in Fig. 4, except $I=2.1\times10^{12}$ W/cm^2. Note the formation of the third plateau and large increase of the height of the second plateau with respect to Fig. 5.

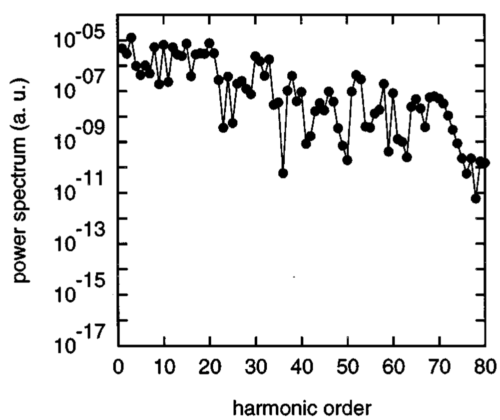

FIG. 8. The same as in Fig. 6, except $E_i=22.3$ eV. Note the merger of the three plateaus, seen in Fig. 7, into a single broad and intense plateau.

magnitude from the previous case (cf. Fig. 5). In addition, one observes the beginning of a *third* plateau from the 70th harmonic upward. The mechanism behind the appearance of the third plateau is completely analogous to that of the second plateau described above and corresponds to an intermediate 31-photon resonance coupling of the incident 5th band with a higher empty band ($\beta = 7$) of the crystal (cf. Fig. 1).

In view of the resonance coupling mechanism between the band of the incident electrons and the empty bands, one expects that the high-harmonic spectra may also be controlled by suitably tuning the energy of the incident electrons. In Figs. 7 and 8 we show the spectra calculated for the incident electron energy $E_i = 22.3$ eV (it corresponds to the Bloch state marked *b* in Figs. 1 and 2). At this energy and $I = 3 \times 10^{-5}$ a.u. (Fig. 7), there appear a second and a third plateau; this is in contrast to the presence of only the second plateau in the corresponding case of Fig. 5. We note that the second plateau is now of shorter extent (from about the 20th to the 30th harmonic) but is nearly three orders of magnitude higher than that in Fig. 5. Finally, at $I = 6 \times 10^{-5}$ a.u. (Fig. 8) the three plateaus seen in the previous figure nearly coalesce into a broad and intense single plateau extending up to the 70th harmonic. This remarkable merging of the plateaus is due to the simultaneous occurrence of several interband multiphoton resonances, which are marked by the empty circles in Fig. 1, involving all the bands from the 3rd to the 7th.

To conclude, we have shown by nonperturbative (Floquet-Bloch) simulations that it is in principle possible to generate very high harmonics via the mechanism of interband resonances, using only moderately intense laser fields that interact with transmission Bloch electrons in a thin crystal. The associated high-harmonic spectra are characterized by the formation of plateaus whose number, height, and range can be controlled by suitably choosing the energy of the transmitting electrons and/or the intensity of the incident laser fields.

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