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Floquet-Bloch theory of photoeffect in intense laser fields

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A Floquet-Bloch analysis of intense-field photoemission of electrons from a model crystal surface is carried out. Simulations of electron emission spectra as a function of the laser intensity show several phenomena, including (a) the appearance of a sequence of emission bands, separated by bands of zero currents, at an interval of the photon energy, (b) a broad modulation of the envelope of the emission spectrum, and (c) the appearance of subbands within an emission band. The results are discussed and interpreted in terms of the field-modified band structure of the system. A comparison of the Floquet-Bloch model and the popular free-electron Sommerfeld model for intense-field photoeffect is made and the difference is shown to be of qualitative significance. [S1050-2947(98)50207-7]

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Recently, photoemission from crystal surfaces subjected to intense laser fields has been an object of vigorous investigations, e.g. [1], motivated both by the desire to understand the behavior of crystals and surfaces in intense laser fields as well as by the need to obtain pulses of high electron currents of very short duration. In particular, it had been found that the interaction of short laser pulses with metal surfaces generates nanosecond or picosecond pulses of highly directional electron beams [2,3]. It is expected that such electron sources can find applications in electron beam lithography and high-resolution electron microscopy [4], as well as in free-electron laser devices [5] and possibly for efficient high harmonic generation [6].

Theoretical investigations of highly nonlinear interaction of laser pulses with crystal surfaces that include the band structure of the system are currently only at their beginning. This is mainly because of the difficulty in the simultaneous nonperturbative treatment of the laser interaction with the band-structure calculation including the surface. Thus, the intense-field surface photoemission problem has been analyzed in the past often within some version of the free-electron Sommerfeld model [7]. For the usual one-photon case, the free-electron model of photoemission could provide an order-of-magnitude agreement with the experimental data on photocurrents [2,7], but in the case of multiphoton photoemission such models failed by many orders of magnitude to give the high currents observed experimentally [2,3].

In this Rapid Communication results of Floquet-Bloch analysis of laser-induced photoelectron emission from a model crystal surface are presented that show a number of phenomena that arise from the electronic band structure *and* its modification due to the laser field. At a given intensity, it is found also to provide orders-of-magnitude larger photocurrents than that given by the usual free-electron models [7].

In Fig. 1 the basic process of emission of electrons from the surface and the alignment of the laser field is indicated schematically. The laser field is assumed to be propagating parallel to the surface, $\hat{\mathbf{k}}_z$, with the polarization vector perpendicular to it, $\hat{\boldsymbol{\varepsilon}}_x$. An electron initially in an occupied band state $\psi_I(x,t)$ inside the crystal (and driven by the laser field) is considered to move toward the surface. After reaching the surface it is either reflected or transmitted with wave functions $\psi_R(x,t)$ or $\psi_T(x,t)$, respectively. In the presence of the laser field, the state $\psi_I(x,t)$ can be most conveniently described by the associated Floquet-Bloch states [8] having positive expectation values of the momenta. The Floquet-Bloch states are generalizations [8] of the usual Bloch states in the absence of the laser field. The reflected wave function $\psi_R(x,t)$ is given by a linear combination of the Floquet-Bloch states having negative expectation values of momenta and the corresponding ‘‘closed-channel’’ states [9]. Similarly, the transmitted wave function in the vacuum $\psi_T(x,t)$ is given by a linear combination of the well-known Volkov states with positive momenta; e.g., [10] and the corresponding closed-channel states that decrease exponentially in the vacuum, away from the surface. The condition of continuity of the total wave function propagating in the crystal and in the vacuum, as well as its space derivative at the surface (for all time t) allows one to determine the desired transmission probability $P_T(\beta,k;v)$, where (β,k) denote the initial band

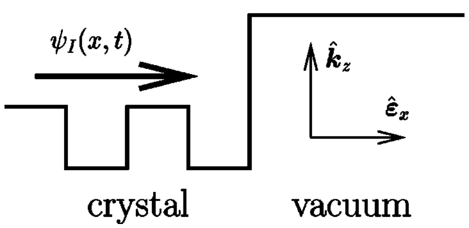

FIG. 1. A schematic description of the photoemission process from a crystal surface. The state of an electron inside the crystal and that in the vacuum are indicated. The laser field is assumed to propagate parallel to the surface, along $\hat{\mathbf{k}}_z$, with the polarization vector perpendicular to it, along $\hat{\boldsymbol{\varepsilon}}_x$.

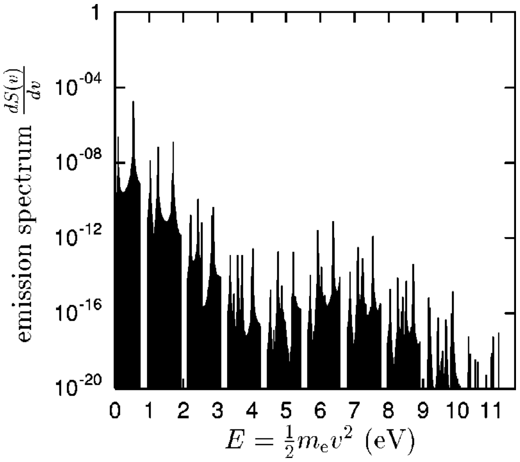

FIG. 2. Above-threshold surface-emission (ATSE) spectrum for the Nd:YAG laser frequency, $\omega = 1.169$ eV, at an intensity $I = 3.51 \times 10^{10}$ W/cm^2. Note a sequence of energy bands separated exactly by the photon energy, sharp resonancelike lines of high currents in the individual emission bands, and a second broad maximum of the envelope between 5 and 9 eV.

index and the Bloch momentum, respectively, and v denotes the final velocity of the photoelectron in the vacuum. The transmission probabilities P_T permit us to define [11] the photoelectron spectrum $dS(v)/dv$,

$$\frac{dS(v)}{dv} = \sum_{\beta,k} \frac{m_e v v_\beta(k)}{4k_F |E'_\beta(k)|} P_T(\beta,k;v), \tag{1}$$

where the summation is over all initially occupied Floquet-Bloch states with positive momenta that fulfill the energy conservation condition

$$E = \frac{1}{2} m_e v^2 = E_\beta(k) \bmod \omega. \tag{2}$$

$v_\beta(k)$ is the expectation value of the velocity of the electron in the initial states (β,k), m_e is the electron mass, k_F is the Fermi momentum, and $1/|E'_\beta(k)|$ is the density of the occupied initial states. For the numerical simulations discussed below, we have used the parameters of a model potential giving a Fermi energy of 5.53 eV and a work function of 5.1 eV [8].

In Fig. 2 we present the photoelectron emission spectrum, calculated for the case of interaction with a neodymium-doped yttrium aluminium garnet (Nd:YAG) laser at a wavelength $\lambda = 1064$ nm and an intensity $I = 3.51 \times 10^{10}$ W/cm^2. The most conspicuous feature of the spectrum seen here is the appearance of a whole sequence of individual energy bands of photoelectrons, separated by strips (white) of zero currents, the edges of which are spaced exactly by the photon energy (1.169 eV). These emission bands are direct analogues of the above-threshold ionization (ATI) peaks, well-known in the case of atomic ionization (e.g., [12]). The separations between the emission bands can appear whenever the photon energy is greater than the width of the initially occupied band. The individual members of such ''above-threshold surface-emission'' (ATSE) spectra also exhibit sharp lines of high electron currents. They are found to be due to the intermediate multiphoton resonances between pairs of Floquet-Bloch bands of the crystal. We may add that similar interband resonances have been predicted also to enhance the signal for high harmonic generation during the passage of electrons through a thin crystal [6]. It can be seen from Fig. 2 that in the low-energy part (up to about 4 eV), the envelope of the ATSE spectrum decreases linearly (in logarithmic scale). This is consistent with the usual power-law behavior (with respect to the photon order) expected from the perturbation theory. However, for larger energies (e.g., between 5 eV and up to 9 eV) we observe the formation of a broad bump in the envelope of the spectrum. This effect becomes more prominent with increasing intensity and can lead to a modulation of the envelope due to the appearance of broad minima and maxima. Such modulations are to be seen in the spectrum of Fig. 3 that is obtained for a higher intensity $I = 1.053 \times 10^{12}$ W/cm^2. A comparison of the positions of the modulation minima in these spectra with the calculated position of the band gaps shows that the former occur only in the vicinity of the latter [8]. The modulation effect, therefore, should be observable in ATSE spectra, independent of the conduction properties of the crystal.

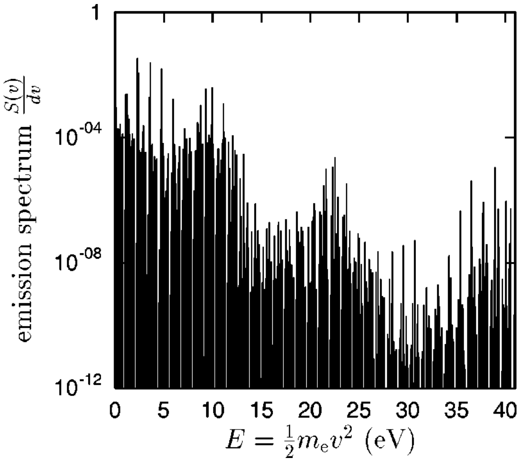

FIG. 3. The same as in Fig. 2, except that $I = 1.053 \times 10^{12}$ W/cm^2.

Under high resolutions a different nonperturbative feature can be seen in the ATSE spectrum. To this end the two lowest above-threshold bands (shown in Fig. 3, corresponding to the intensity $I = 1.053 \times 10^{12}$ W/cm^2) in the energy interval up to 2 eV, are presented in a magnified scale, in Fig. 4 (lower panel). Also shown are the Floquet-Bloch bands and their replicas in the same energy interval (upper panel). The portions of the bands and their replicas that correspond to the initially occupied (positive velocity) states are marked bolder in the upper panel. We note that due to the presence of multiphoton resonances at specific values of the quasimomenta k, there appear a number of avoided crossings that create the minigaps seen in the band structure (upper panel). A comparison of the energies at which the minigaps within the occupied states arise (upper panel) with the positions of the minigaps in the emission spectrum (lower panel) shows an exact *one-to-one* correspondence between them. This correspondence provides, therefore, a way of observing the minigaps in the field-modified band structure of a crystal, by high-resolution measurements of the ATSE current [13]. It should, however, be noted that all the structures or substructures predicted here may not be resolved by usual experi-

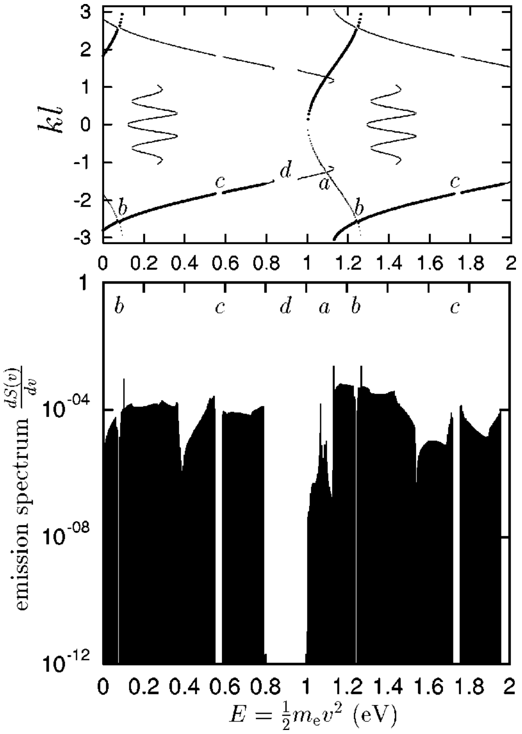

FIG. 4. Magnification of the two lowest above-threshold bands, at $I=1.053\times10^{12}$ W/cm^2, shown in Fig. 3 (lower panel). A number of minigaps are to be noticed in the bands (upper panel). The portions of the initially occupied (positive velocity) states are marked by the darker parts. A *one-to-one* correspondence between the positions of the minigaps (upper panel) and the fine structures of zero currents (lower panel) is also seen to be replicated at an interval of the photon energy.

ments with polycrystals at ordinary conditions when the collisional incoherence time τ is very small $[2.9\times10^{-14}$ sec (e.g., p. 401 [14])], but would require more stringent conditions as, for example, in the case of cyclotron resonance experiments for measuring the Fermi surface of a metal, e.g., with highly pure single crystals at low temperatures, when τ can be much larger $[\tau\approx10^{-11}$ sec at 4 K (p. 537 [14])]. It would be also necessary, in order to avoid high-space-charge formation that can distort the spectra, to use short pulses (e.g., between 0.1 to 1 picosec) and/or peak intensities sufficiently [15] below the saturation intensity; the latter intensity has been estimated to be of the order of 3.5×10^{12} W/cm^2, at the Nd:YAG frequency [6]. We point out further that use of a shorter-wavelength laser, e.g., a Ti:sappphire ($\lambda=780$ nm) or a KrF laser ($\lambda=248$ nm), would increase the separation between the ATSE bands and hence the width of a zero-current interval, permitting one to use less stringent electron-energy resolution for their detection.

Finally, in Fig. 5, the photoelectron spectrum obtained from the Floquet-Bloch theory (lower panel) is compared with that obtained from the corresponding free-electron Sommerfeld model (upper panel) at $I=3.51\times10^{11}$ W/cm^2 and $\omega=1.169$ eV. It is seen that they differ greatly in their structures; e.g., the gaps between the emission bands and the modulation of the envelope are clearly absent in the free-electron model. Furthermore, the free-electron model greatly underestimates the emission currents at all energies; e.g., by four orders of magnitude for the lowest energies and by more than 15 orders of magnitude for electrons above 10 eV. Clearly, the influence of electronic band structure cannot be neglected in modeling the photoelectron emission current in intense laser fields.

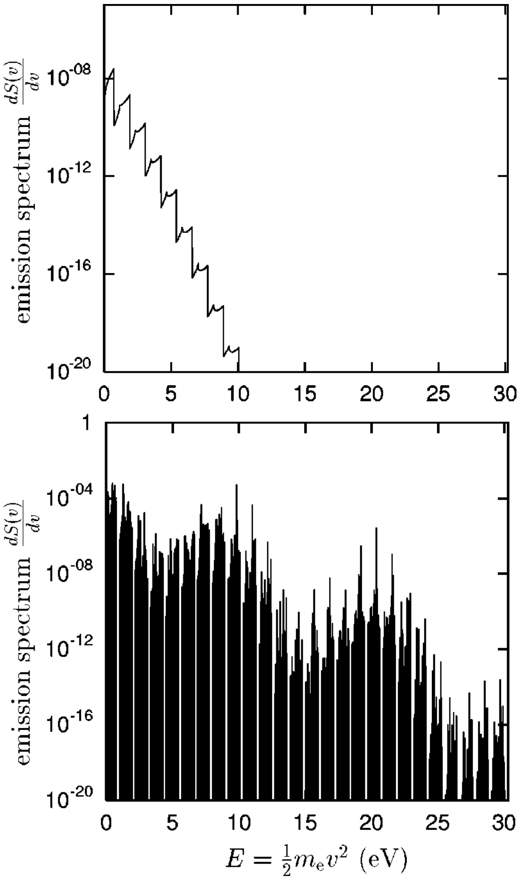

FIG. 5. Comparison of photoelectron emission spectrum: Floquet-Bloch theory (lower panel) and the free-electron Sommerfeld model (upper panel); $\omega=1.169$ eV and $I=3.51\times10^{11}$ W/cm^2.

To conclude, a Floquet-Bloch analysis of the interaction of intense laser light with a model crystal surface is presented. Numerical calculations of the photoelectron spectrum reveal a sequence of emission *bands*, separated by strips of zero current at an interval of the photon energy. They constitute a kind of above-threshold surface-emission (or ATSE) spectrum that is analogous to the so-called above-threshold ionization (ATI) spectrum, well-known in atomic physics. Distinct ATSE bands are found to occur whenever the photon energy is greater than the width of the initially occupied electronic band of the system. The individual ATSE bands also show sharp lines that arise from multiphoton interband resonances. At high intensities, the envelope of the ATSE spectrum is found to be *modulated* by broad minima and maxima, as a function of emission energy. Under high resolution, the individual emission bands can reveal subbands that also are replicated at an interval of the photon energy. These subbands are shown to be correlated *one to one* with the appearance of minigaps in the field-modified band structure of the surface, and thus can provide a means of detecting the presence of the latter. A comparison is made between the prediction of the present theory and that of the corresponding free-electron Sommerfeld model. It is found that the presence of the electronic band structure of the surface can greatly enhance both the height and the range of the emission spectrum, compared to that predicted by the free-electron model.

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