Resonant generation of high harmonics

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A mechanism for the resonant generation of high harmonics of a laser beam is discussed. This method may make it possible to increase the conversion coefficient substantially and to move to shorter wavelengths. In particular, it is estimated that harmonics in the region $\lambda \simeq 100$ Å can be generated with an efficiency on the order of 10^{-5} in a beryllium plasma. © *1994 American Institute of Physics.*

Generating high harmonics of laser light, i.e., harmonics whose indices run to two and three digits and whose wavelengths lie in the x-ray part of the spectrum, in atomic jets[1] and low-density plasmas[2] is a promising method for producing coherent x radiation. This phenomenon was first observed[1] in the late 1980s. It occurs in fields comparable to intraatomic fields, along with an ionization of the atoms and ions which are generated. Record-short wavelengths near 8 nm have been achieved. Unfortunately, the conversion coefficients which have been realized so far are very small, about 10^{-10} into one harmonic in this region.[3]

In this letter we wish to discuss a version of resonant high-harmonic generation which may make it possible to raise the conversion coefficients and to move to shorter wavelengths, in particular, to the "water window," 20–42 nm. The idea proposed below was stimulated by the results of a numerical study of the Schrödinger equation for a hydrogen atom in the strong high-frequency electric field of a light wave ($I = 1.75 \times 10^{14}$ W/cm^2, $\hbar\omega = 0.4$ Ry), carried out by DeVries.[4] It can be concluded from an analysis of these results that in the well-developed stage of the interaction, in which the average electron energy exceeds the ionization energy, an electron continues to oscillate near the nucleus for many optical periods, preserving the relatively small dimensions and radiating harmonics. In the case of many-electron atoms and ions, the interaction with the electrons of the relatively "porous" atomic core should prevent large-amplitude (quasi-) periodic motion of an electron near the parent ion. It seems improbable even in low-frequency fields. The discussion below is based on the assumption that a motion of this sort is possible in the case of lithium-like systems.

A lithium-like ion with a charge $Z-1$ can be thought of, somewhat crudely, as a system consisting of two subsystems: a compact, strongly bound helium-like ion of charge Z and an outer electron, bound relatively weakly to the compact ion and lying far away from it. This outer electron plays the role of an antenna. Oscillating in the optical field, the outer electron not only radiates harmonics but also induces radiation of harmonics by K-shell electrons. Resonances of the helium-like ion should naturally be manifested in this radiation. At the same time, they would have little effect on the motion of the outer electron. This motion can be thought of as motion in the field of a structureless (point) charge.

Since the K shell has small dimensions, the field produced by the outer electron at the position of the helium-like ion, $\mathbf{E}_{ei}$, can be assumed to be nearly uniform. We denote by $\mathbf{E}_j$ the amplitude of the jth harmonic of this field. For $j \neq 1$, the amplitude of the dipole moment of the helium-like ion at the frequency $j\omega$ is $\mathbf{d}_{ij} = \alpha_i(j\omega)\mathbf{E}_j$, where α_i is the polarizability of the ion. Acting on the outer electron, in addition to the optical field, is the field set up by the helium-like ion. We denote by $\mathbf{E}_{ie}$ the average value of this field, and by $-e\mathbf{E}_{ie}$ the force acting on the outer electron. Comparing the latter force with the force $Ze\mathbf{E}_{ei}$, acting on the helium-like ion, and using Newton's third law, we find $\mathbf{E}_{ie} = Z\mathbf{E}_{ei}$. Correspondingly, the amplitude of the jth harmonic of the field $\mathbf{E}_{ie}$ is $Z\mathbf{E}_j$, while the amplitude of the jth harmonic of the dipole field at the outer electron in the case $j \neq 1$ is $\mathbf{d}_{ej} = Z\alpha_e(j\omega)\mathbf{E}_j$, where $\alpha_e(\omega) = -e^2/m\omega^2$ is the polarizability of an electron. The amplitude of the resultant dipole moment of the lithium-like ion is thus

$$\mathbf{d}_j = [Z\alpha_e(j\omega) + \alpha_i(j\omega)]\mathbf{E}_j. \tag{1}$$

At resonant frequencies, the ion component of the resultant dipole moment in (1) can clearly be predominant. This component has been ignored in previous studies of high-harmonic generation.

Yet another resonant factor arises in the expression for the harmonic intensity because of the dispersion of the refractive index, $n(\omega)$. We assume that the generation occurs in a plane plasma slab on which light is incident normally (along the x axis). The plasma contains only lithium- and helium-like ions, with respective densities N_{Z-1} and N_Z, and free electrons, with a density $N_e = ZN_Z + (Z-1)N_{Z-1}$. Since the average energy of the outer electron in a lithium-like ion is considerably larger than the ionization energy, its motion can be treated as nearly free in a calculation of the refractive index. Assuming that the polarizability of the helium-like ion is small except at the frequencies of resonant harmonics, and assuming that the polarizability of the electron is small except at the fundamental frequency, we write

$$n(\omega) - n(j\omega) \simeq 2\pi(N_Z + N_{Z-1})[Z\alpha_e(\omega) - \alpha_i(j\omega)]. \tag{2}$$

If the slab is sufficiently thin, we can ignore the change in the intensity of the exciting light in the slab, and we can also ignore geometric effects which influence the synchronization of the high-harmonic generation.[5] In this case we can treat it in the plane-wave approximation. Using (1) and (2), and setting $N_{Z-1}(x)/N_Z(x) = \text{const}$, we find the following expression for the conversion coefficient $\eta_j = I_j/I$ (I_j is the intensity of the harmonic):

$$\eta_j = |E_j/E|^2 \theta_j N_Z^2 (N_Z + N_{Z-1})^{-2} |1 - e^{i\psi_j}|^2, \tag{3}$$

where $\psi_j = j(\omega/c)\int(n(\omega) - n(j\omega))dx$, $E = (2\pi I/c)^{1/2}$, and

$$\theta_j = |Z\alpha_e(j\omega) + \alpha_i(j\omega)|^2 / |Z\alpha_e(\omega) - \alpha_i(j\omega)|^2. \tag{3a}$$

The case of a relatively precise resonance, $j\omega \approx \omega_i$, where ω_i is one of the resonant frequencies of the helium-like ions, is naturally the most interesting case from several standpoints. In principle, one could also raise the question of creating a population inversion on their transitions in this case. However, that question requires a special analysis. We assume here that the detuning $\hbar\Delta = \hbar(j\omega - \omega_i)$ is on the order of 1 eV. We then have

$$\theta_j \simeq |\alpha_i(j\omega)/Z\alpha_e(\omega)|^2 \simeq |f\omega/2Zj\Delta|^2, \tag{3b}$$

where f is the oscillator strength of the resonant transition.

To evaluate possible values of $\mathbf{E}_j$, we adopt several additional assumptions.

1. We denote by $\varphi$the wave vector of the outer electron, and by $\mathbf{r} = \int \boldsymbol{\rho} |\varphi(\boldsymbol{\rho})|^2 dV$ its "radius vector." The field $\mathbf{E}_{ei}$ is then directed along $\mathbf{r}$, is smaller in absolute value than e/r^2, and can thus be written in the form

$$\mathbf{E}_{ei} = e\mathbf{r}/(r^2 + r_0^2)^{3/2}, \tag{4}$$

where $r_0(t)$ is a positive and otherwise unknown parameter.

2. The radius vector $\mathbf{r}(t)$ varies nearly harmonically:

$$\mathbf{r} = \mathbf{r}_e \sin(\omega t + \vartheta), \tag{5a}$$

$$\dot{r}_e \ll \omega r_e > 0, \quad \dot{\vartheta} \ll \omega. \tag{5b}$$

3. We assume $t_k = (k\pi - \vartheta)/\omega$ and $\tau_k = r_0(t_k)/\omega r_e$, where k is an integer. On the interval $(t_k - \tau_k,\ t_k + \tau_k)$ the following conditions then hold:

$$r_0 \ll r_e, \quad |r_0 \dot{r}_0| \ll \omega r_e^2. \tag{6}$$

4. Finally, the quantity $r_0(t_k)$ varies only slightly with a variation in the index k:

$$r_0(t_k) - r_0(t_{k+1}) \ll r_0(t_k). \tag{7}$$

Condition (5a) allows us to write field (4) in the form

$$\mathbf{E}_{ei} = \sum \mathbf{E}_j e^{-ij\omega t} + \text{c.c.},$$

where $E_{2N} = 0$. Under condition (5b) with $N > 0$, we can also write

$$|E_{2N+1}|^2 \simeq \frac{4e^2(N+1)}{\pi r_e^3 r_0} \left[1 - \frac{r_e}{2r_0} \left(1 - \sqrt{\frac{N}{N+1}} \right) \right]^2 \frac{(1 - 2r_0/r_e)^2}{(1 - 2r_0^2/r_e^2)^2}. \tag{8}$$

[Expression (8) cannot be used if the absolute value of the difference in square brackets is much less than one. To find it, we should expand (4) in a Taylor series in $\cos 2\omega t$, express the sine and cosine functions in terms of exponential functions, and sum the coefficients of $e^{ij\omega t}$.]

Because of (6), the field $\mathbf{E}_{ei}$ and therefore its spectrum are nearly independent of the values of $r_0(t)$ far from the points t_k. They remain essentially the same if the actual $r_0(t)$ dependence is replaced by a broken line connecting points $[t_k, r_0(t_k)]$ on the t, r_0 plane. We will understand $r_0(t)$ below to mean specifically this sort of function. Inequali-

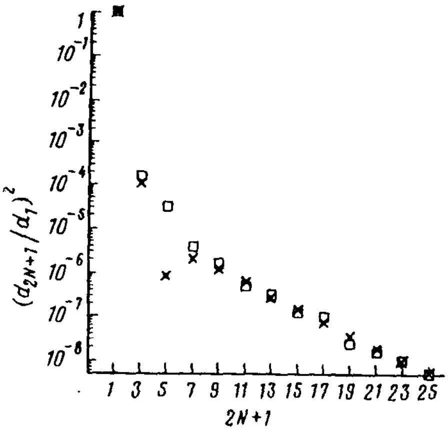

FIG. 1. Values of d_j^2/d_1^2 calculated for a hydrogen atom from Eqs. (1), (8), and (9) with $r_0=r_B$ (crosses) and as given in Ref. 4 (squares).

ties (5b), (6), and (7) then guarantee that the quantities $\mathbf{E}_j$ are weak functions of the time and allow us to assume that they are the Fourier amplitudes of the field $\mathbf{E}_{ei}$ in (3).

Calculating $\mathbf{E}_1$, we find an equation which relates r_e and r_0:

$$r_e \simeq [(8\pi I/c)^{1/2} + 2Ze(1-2r_0/r_e)/(\pi r_e r_0^3)^{1/2}]e/m\omega^2. \tag{9}$$

Using (1), (8), (9), and $d_1^2=e^2r_e^2/4$, and varying the parameter r_0, we calculated values of d_j^2/d_1^2 for the conditions of the numerical calculations of Ref. 4, and we compared the results with the results given in Ref. 4. The best agreement was found with r_0 equal to the first Bohr radius r_B (Fig. 1). In this case, expression (8) is not valid for the fifth harmonic (see the discussion above). For the seventh, it yields a result which is half that in Ref. 4. For the other harmonics, the results differ by a factor of no more than 1.5. (The values of d_j^2/d_1^2 themselves vary from about 1.5×10^{-4} to 5×10^{-9} as j is varied from 3 to 25.)

The good agreement of the results indicates that the dimensions of the distribution $|\varphi(\boldsymbol{\rho})|^2$ under the conditions of Ref. 4 track the changes in the position of its center of gravity almost adiabatically. It is natural to suggest that the approximate equality $r_0 \simeq Zr_B \mathrm{Ry}/U$ also holds in a lithium-like ion, where U is the ionization potential, if the adiabatic parameter $\gamma=(2mU)^{1/2}\omega/|2eE|$ is sufficiently large (it is $\gamma\simeq 2$ under the conditions of Ref. 4). At frequencies of the exciting light which are technically feasible, the condition $\gamma>1$ can be satisfied, along with the condition of rapid ionization, for a lithium atom and for lithium-like beryllium. To illustrate the situation we will go through the calculations for beryllium, ignoring the Stark shift of the levels.

We assume that we are using the beam from a KrF excimer laser. Within its gain band, we can adjust to the frequency $\omega\simeq(\omega_{\mathrm{Be}}+0.5\ \mathrm{eV}/\hbar)/25$, where $\hbar\omega_{\mathrm{Be}}=123.67$ eV is the energy of the $1^1s\to 2^1p$ transition in helium-like beryllium. At an intensity $I\simeq 6\times 10^{14}$ W/cm^2, values $\gamma\simeq 1.3$ are realized, and the value of the exponential function in the Keldysh formula for the ionization probability is approximately the same as under the conditions of Ref. 4. Working from Eqs. (8) and (9) with these parameter values and $r_0=Zr_B\mathrm{Ry}/U\simeq 0.79$ Å , we find $|E_{25}/E|^2\simeq 2.4\times 10^{-3}$. At $f\simeq 0.276$ [as in the helium atom; see (3b)] we have $\theta_{25}\simeq 0.75\times 10^{-3}$, and the maximum value of the conversion coefficient in (3) is $\eta_j \simeq 0.8 \times 10^{-5}$. This value is more than four orders of magnitude higher than the energy conversion coefficient which has been achieved in high-harmonic generation in this frequency range.[3]

Although it is unlikely that the equality $r_0 \simeq Z r_B \mathrm{Ry}/U$ will be satisfied at technically accessible frequencies for ions of heavier elements, conditions (5)–(7) can apparently also be satisfied for ions of boron and carbon. The scheme discussed above should then lead to a relatively high generation efficiency. Even the resonant transitions of helium-like carbon lie in the x-ray "water window." Some of them agree well with frequencies of high-power lasers and hold promise for the realization of resonant high-harmonic generation.

Thin plasma slabs with a controllable ion composition can be produced by (for example) evaporating films with a thickness on the order of 10^{-6} cm with short laser pulses.

[1] A. McPherson *et al.*, J. Opt. Soc. Am. B **4**, 595 (1987); M. Ferray *et al.*, J. Phys. B **21**, L31 (1988).
[2] S. Kubodera *et al.*, Phys. Rev. A **48**, 4576 (1993).
[3] A. L'Huillier and Ph. Balcou, Phys. Rev. Lett. **70**, 774 (1993).
[4] P. L. DeVries, J. Opt. Soc. Am. B **7**, 517 (1990).
[5] A. L'Huillier *et al.*, J. Phys. B **24**, 3315 (1991).

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