X-ray third-harmonic generation in plasmas of alkalilike ions

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We demonstrate theoretically the feasibility of x-ray near-resonant third-harmonic generation in a number of plasmas of Li- and Na-like ions as well as third-harmonic generation enhanced by phase matching using buffer plasmas.

Increasing availability of x-ray lasers (XRL's) makes research on coherent x-ray nonlinear optics a timely endeavor.[1] Recently absorption saturation,[2,3] nonlinear refractive index,[3] and four-wave mixing[4] have been considered. In this Letter we address the x-ray third-harmonic generation (THG), which is important as one of the ways of producing shorter-wavelength coherent radiation.

Plasmas in which the x-ray THG could in principle be similar to the optical THG in gases or vapors are the most natural media candidates for such a process. Although the main elements of the theory of THG in gases and plasmas are well known, the major problem in the x-ray case is identifying appropriate ions and plasmas that would resonantly match the available XRL's. Since dipole moments of transitions in general decrease with the wavelengths, significant conversion efficiency can be expected only for a near-resonant THG. It is well known[5] that the best resonant conditions for THG are (i) the presence of a two-photon resonance from the initial (usually ground) level g to a certain level (level b in Fig. 1) that is optically inaccessible otherwise and (ii) the presence of two other levels (a and c in Fig. 1) that are nearly resonant to the fundamental ω and the third 3ω harmonics, respectively, and strongly optically coupled with both initial g and two-photon b levels. The desirable conditions are that the resonance gb be as exact as possible, whereas the detunings for the resonances $ga \to \omega$ and $gc \to 3\omega$ ideally are to be slightly greater than the width of the respective transition (in order to avoid strong absorption in the closer vicinity of exact resonances). Since no tunable XRL is available now, one has to look for plasmas that may allow for resonances to some of the ~40 existing XRL lines.[1] This search is complicated by insufficient information on energy levels and transition probabilities of ions. In this respect, plasmas of alkalilike ions present perhaps the best opportunity because of readily available atomic data.[6,7]

In this Letter we demonstrate the feasibility of the x-ray THG by some available XRL lines resonantly coupled with plasmas of Li- and Na-like ions. Identified resonant couples, XRL line/plasma transition, and estimated conversion efficiencies are presented in Table 1. Owing to the lack of sufficiently accurate information on XRL wavelengths, transition probabilities, and plasma conditions, our quantitative results should be viewed as preliminary estimates. By the same token, at this stage, some of the competing processes such as multiphoton absorption and Stark effect have also been neglected.

We use standard results of the theory of THG in gases and vapors,[5,8,9] modifying them mainly to account for the specifics of the plasma dispersion. The x-ray refractive index due to free plasma electrons is $n_p(3\tilde{\nu}) \simeq 1 - \omega_p^2/\omega^2$,[10] where ω is the frequency of the incident XRL radiation, $\omega_p = (N_e e^2/\epsilon_0 m)^{1/2}$ is the plasma frequency, and N_e is the plasma electron density. For all but one of the couples in Table 1, this free-electron component of the refractive index is much larger than the resonant (bound-electron) one, which can therefore be neglected. We also assume that the plasma is homogeneous enough for us also to neglect the refraction owing to inhomogeneous electron distribution across the laser beam. As a result, the phase mismatch $\Delta k_p = 6\pi\lambda^{-1}[n_p(3\omega) - n_p(\omega)]$, where $\lambda = 2\pi c/\omega$, is positive (which prevents one from using too tight focusing of the XRL beam, which in such a case may inhibit THG[8]). Thus we assume loose focusing of the XRL beam, $L_d \gg L$, where $L_d = 2\pi w_0^2/\lambda$ is the diffraction length of the beam, w_0 is the beam radius at the waist, and L is the length of the plasma column. Conversion efficiency $C_{\text{eff}} \equiv P^{(3)}/P$ [P and $P^{(3)}$ are the power of the fundamental and the third harmonic, respectively] can then be written as[9]

$$C_{\text{eff}} = (3\pi^2/4\lambda^2\epsilon_0^2c^2)\,|\chi_{\text{THG}}|^2 N_i I^2 L^2 \,\text{sinc}^2(\Delta k L/2) \quad (1)$$

(in SI units). Here $I = P/A$ describes the intensity of the XRL radiation, $A = L_d\lambda/4$ is the effective area of the Gaussian beam, N_i is the number density of the ions of interest (we will refer to N_i as to ion density), and $\text{sinc}(x) \equiv x^{-1}\sin(x)$. The third-harmonic susceptibility χ_{THG} in Eq. (1) in the case of linear polarization of the fundamental harmonic is as

$$\chi_{\text{THG}} = \frac{1}{\hbar^3\epsilon_0}\sum_{abc}\frac{\mu_{ga}\mu_{ab}\mu_{bc}\mu_{cg}}{(\omega_{cg} - 3\omega)(\omega_{bg} - 2\omega)(\omega_{ag} - \omega)}, \quad (2)$$

where ω_{ik} and μ_{ik} are the frequencies and the dipole

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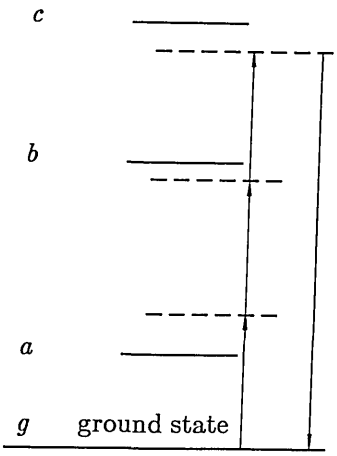

Fig. 1. Typical energy-level diagram for near-resonant THG. Only one of the possible level combinations is depicted.

moment matrix elements of the transitions between the levels of interest, respectively. Note that there is no summation over the g index, since we have excluded weighting over initial states, assuming that almost all the ions of interest in the unperturbed plasma are at the only nondegenerated ion ground level g so that $E_g = 0$ and $\omega_{ig} = E_i/hc$, where $i = a, b, c$ and E_i are the energies of the levels. Theoretically, the summation in Eq. (2) includes all the ion excited levels. Usually, however, only a few resonant level combinations dominate, and the rest can be neglected. General expressions for C_{eff} and χ_{THG} include the laser linewidth and the widths of the transitions.[11] The coherence of the existing XRL's is far from perfect [the relative linewidth of Se XRL 20.6- and 20.9-nm lines is $\simeq 2 \times 10^{-4}$ (Ref. 12)]. However, for all the couples in Table 1 detunings are larger than 10^{-3} (except for the third one, where some of the detunings are only slightly larger than 2×10^{-4}). Therefore our estimates are not significantly affected by insufficiently high coherence of existing XRL. Following the method of Ref. 11, the finite widths of all the resonant transitions in Eq. (2) are also neglected, since they are significantly smaller than detunings for all the couples in Table 1.

Direct calculations of μ_{ik} require knowledge of the wave functions of the ions and are not available in most of the cases. Instead, following Ref. 12, we express the μ_{ik}'s through the transition probabilities A_{ik} as $\mu_{ik} = s_{ik}(\pi h c^3 A_{ik}\epsilon_0/2\omega_{ik}^3)^{1/2}$, where the signs $s_{ik} = \pm 1$ for a Li (Na)-like ion are assumed the same as for the Li (Na) atom in Ref. 8. The maximum value of C_{eff} for a given mismatch Δk is achieved at $L = L_{\max} = \pi/\Delta k$ or $L_{\max}(\text{cm}) \simeq 4.2 \times 10^{19}\lambda^{-1}(\text{nm})N_e^{-1}(\text{cm}^{-3})$. With all these assumptions, we can rewrite Eq. (1) as

$$(C_{\text{eff}})_{\max} \simeq 0.003M^2(N_i/N_e)^2I^2, \tag{3}$$

with

$$M = \lambda^{-2}\sum_{abc} s_{abc} \times \frac{[E_a(E_b - E_a)(E_c - E_b)E_c]^{-3/2}(A_{ag}A_{ba}A_{cb}A_{cg})^{1/2}}{(E_c - 3\lambda^{-1})(E_b - 2\lambda^{-1})(E_a - \lambda^{-1})}, \tag{4}$$

where the XRL radiation intensity is in watts per squared centimeter, the energy of the levels is in inverse centimeters (as is usual in spectroscopic tables), λ is in centimeters, and $s_{abc} = s_{ga}s_{ab}s_{bc}s_{cg}$. As a result of plasma dispersion, C_{eff} does not depend on the plasma density, although L_{max} depends on N_e; e.g., for a low N_e the length L_{max} would become unrealistically large.

Table 1. X-Ray THG in Plasmas[a]

Resonant Couples XRL line λ (nm) / Plasma/E_{ion} (eV)	$(\lambda/3)$ (nm)	I (W cm^{-2}) for $C_{\text{eff}} = 10^{-8}$	Buffer ions α/E_{ion}'(eV)	I (W cm^{-2}) for $C_{\text{eff}} = 10^{-6}$
Ge^{22+}28.646 / K IX/176	9.5487	8×10^{12}		
Se^{24+}20.638 / Sc XI/250	6.8793	2×10^{14}		
Se^{24+}18.243 / Ne VIII/239	6.0810	5×10^{12}	Si VII / 0.06/247	2×10^{11}*
C^{5+}18.2097 / Ne VIII/239	6.0699	2×10^{13}	Be IV / 0.20/218	10^{10}
Y^{29+}15.50 / V XIII/337	5.167	2×10^{14}		
Ag^{37+}9.93 / Al XI/442	3.31	5×10^{14}		
Eu^{35+}6.583 / Ga XXI/807	2.194	2×10^{14}	Ti XIV / 0.06/863	7×10^{10}
Na^{10+}5.4194 / Cl XV/809	1.8065	10^{15}		

[a] α is the ratio of the required buffer-ion density to the electron density, and E_{ion} and E_{ion}' are the ionization potentials of the main and buffer plasmas, respectively. The asterisk denotes the result obtained for an ion density of 10^{18} cm^{-3}.

The results of calculations based on Eqs. (3) and (4) can be found in Table 1 in terms of the XRL intensities required to attain $C_{eff} \simeq 10^{-8}$. They show that the observable (although yet relatively weak) THG intensities can be attained by available XRL's. (For example, the power of Ge^{22+} XRL required for $C_{eff} \simeq 10^{-8}$ in the loose focusing limit can be estimated as several megawatts, which is close to the peak power of the existing XRL. It corresponds to the input energy of several hundreds of microjoules in a 100-ps pulse). One of the ways to improve the x-ray THG is to facilitate ideal phase matching condition $\Delta k = 0$. Similarly to the optical THG,[8] this can be done by adding a buffer medium or, in our case, buffer ions. Let some transition from the ground level of the buffer ions be resonant to the third harmonic, $\lambda/3 < \lambda_0$, where λ_0 is the central wavelength of the transition, and let no buffer-ion transition be in close resonance to the fundamental harmonic. Then the resonant refraction by buffer ions may compensate for free-electron phase mismatch if

$$n_p(3\omega) - n_p(\omega) \simeq 1 - n_{buf}(3\omega), \quad (5)$$

where n_{buf} is the index of refraction due to buffer ions. Following Ref. 3 and assuming that (i) the Doppler broadening of the buffer transition is much larger than the homogeneous broadening, (ii) the upper level of this transition is almost empty, and (iii) the generated third harmonic is weak enough for one to neglect saturation effects, one can write

$$1 - n_{buf}(3\omega) \simeq 2 \times 10^{-3}(\lambda/3)(f/\Delta\nu^D)N_i'\overline{w}(x), \quad (6)$$

with the buffer-ion density N_i' in inverse centimeters cubed and λ in centimeters. Here f and $\Delta\nu^D$ are the oscillator strength and the Doppler width of the buffer transition, respectively; $\overline{w}(x) \equiv \mathrm{Im}\, w(x + iy)$ at $y = 0$ with $w(x + iy)$ being the complex error function[13]; and $x = 2\sqrt{\ln 2}\,(3\nu - \nu_0)/\Delta\nu^D$. Relations (5) and (6) yield the following condition for $\Delta k = 0$:

$$(N_i'/N_e)\overline{w}(x) \simeq 4 \times 10^{-4} f^{-1}[T_i'(\mathrm{eV})/M_i'(\mathrm{a.m.u.})]^{1/2}, \quad (7)$$

where T_i' and M_i' are the buffer-ion temperature and mass, respectively. In addition, the ionization potentials of active and buffer plasma should not differ substantially so that significant abundances of both ions could exist at the same temperature. Some of possible choices of buffer plasmas are presented in Table 1 (corresponding atomic data can be found in Refs. 14–18). For $\Delta k = 0$, Eq. (1) yields

$$C_{eff} \simeq 4.5 \times 10^{-28}\lambda^2 M^2 N_i^2 L^2 I^2, \quad (8)$$

where N_i is in inverse centimeters cubed and L is in centimeters. For $N_i \simeq 10^{20}$ cm^{-3} (10^{18} cm^{-3} for the Se^{24+} 18.243-nm line, to avoid in this case strong resonant third-harmonic absorption) and $L = 30$ cm, Eq. (8) gives the conversion efficiency (Table 1) enhanced by many orders of magnitude compared to the efficiency of unbuffered THG. Such enhancement may be ascribed to the increase of L from L_{max} (which would be in the micrometer range for such dense plasmas) to 30 cm made possible by buffer plasma. Required plasma conditions may conceivably be attained in some discharge devices.[19,20]

It is worth noting that for C^{5+} XRL radiation in Ne VIII plasma one may expect the phase mismatch to be substantially compensated by the resonant refraction at the third harmonic without buffer ions.

Further significant enhancement of the efficiency (up to several orders of magnitude for the same XRL intensity) requires a search for better resonant couples of XRL line/plasma. Resonant conditions for THG can also be enhanced by using ion beams in which the resonant tuning can be controlled by the Doppler effect. An alternative scheme of x-ray THG, by using the resonance with the bottom of the photoionization continuum of appropriate ions, is the subject of continuing research.

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