X-ray laser frequency near-doubling and generation of tunable coherent x rays in plasma

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We identify and evaluate plasmas in which efficient x-ray laser frequency near-doubling is expected for a number of available x-ray lasers by means of four-wave mixing with Nd or KrF lasers. In some of these plasmas, four-wave mixing of coherent x rays and tunable optical radiation may result in tunable coherent x-ray radiation powerful enough for x-ray laser spectroscopy.

In spite of rapid progress in x-ray laser (XRL) research,[1] the availability of coherent x-ray sources is still very limited, especially at shorter wavelengths. In the visible, IR, and UV domains, the variety of coherent radiation sources is greatly enhanced by nonlinear optical transformations of laser radiation. Development of a number of quite powerful XRLs[1] makes one suggest that nonlinear optics may be useful in generating coherent radiation at new, in particular, higher frequencies in this soft-x-ray domain as well. Recently, the feasibility of x-ray third-harmonic generation in plasma has been theoretically demonstrated.[2] Another third-order nonlinear effect, which may lead to x-ray laser frequency upconversion, is four-wave mixing of coherent x-ray and optical radiation.[3] One could expect quite high conversion efficiency for this process because (i) powerful optical lasers may be used; (ii) one of the participating frequencies is relatively low (optical); (iii) close resonances are more likely. Moreover, this process may appear instrumental in generating tunable coherent x rays.

General theoretical consideration of short-wavelength four-wave mixing in plasma can be found in Ref. 3. In the present letter, we identify and evaluate plasmas in which efficient x-ray laser frequency near-doubling is expected for a number of available x-ray lasers by means of resonant four-wave mixing with Nd or KrF lasers. In some of these plasma, four-wave mixing of coherent x rays and tunable optical radiation may result in tunable coherent x-ray radiation powerful enough for x-ray laser spectroscopy.

Of the various four-wave mixing processes, we have chosen to study difference-frequency mixing, since in this case it seems relatively easy to attain optimal phase matching. As holds for x-ray nonlinear optics in general, only resonantly-enhanced effects may be observed at the present level of x-ray laser technology. We are therefore interested in resonant nonlinear media (plasmas) for the following difference-frequency mixing process:

$$\omega = 2\omega_{\rm XRL} - \omega_{\rm opt}. \qquad (1)$$

Here, $\omega_{\rm XRL}$ is the frequency of an XRL, and $\omega_{\rm opt}$ is the frequency of an optical laser. For the purpose of XRL frequency near-doubling, we consider processes (1) with two particular optical lasers: a powerful Nd laser, which is used to pump most XRLs, or a very bright, short-pulse KrF laser. (In the latter case, the converted x-ray pulse duration may be much shorter than the duration of the incident x-ray pulse.) If a *tunable* optical laser is used instead, the result is tunable coherent x-ray radiation at almost doubled XRL frequency.

Ions of two isoelectronic sequences appear to be most useful for x-ray four-wave mixing. For many C-like ions (Fig. 1), the energy of $2s^2 2p^2\ ^3P$–$2s2p^3\ ^3P^0$ and $2s2p^3\ ^3P^0$–$2p^4\ ^3P$ transitions are very close to each other. This may be useful in providing one- and two-x-ray photon resonances. Transitions from $2p^4\ ^3P$ levels to the closest lower level $2s2p^3\ ^3S_1$ correspond to x-ray radiation if the previous two do, and therefore, no three-photon resonances are possible for processes (1) in C-like ions. By the same token, tunable x-ray radiation may be generated in these ions through process (1) for a broad range of optical frequencies with almost uniform efficiency.

Na-like ions may provide all three resonances for processes (1). Also, the plasma temperature required for the substantial presence of the Na-like ionization stage is relatively low due to low ionization potentials of Na-like ions. A disadvantage of Na-like ions as x-ray four-wave mixing nonlinear media is that initial levels of nonlinear transitions should be excited levels (only then are simultaneous one- and two-photon resonances possible), and it may be difficult to populate these levels significantly.

Resonant combinations of lasers and ions, with estimates of conversion efficiency, are listed in Table I. The tight-focusing limit $b \ll L$ is assumed (b is the confocal parameter and L is the length of a plasma cell). To evaluate the minimum plasma length required, we make use of the reported[4] spot size $\omega \simeq 2\ \mu$m of the Y^{29+} 155 Å XRL (confocal parameter $\simeq 0.15$ cm). Similar values of the confocal parameter are readily attainable for the Nd laser fundamental harmonic. We assume $b = 0.15$ cm for all the participating beams. It means that plasma length $L_{\min} = 10\ b \simeq 1.5$ cm satisfy the tight-focusing condition.[5]

The general expression for the conversion efficiency $C_{\rm eff}$ of the difference-frequency generation[6] may be rewritten as follows:

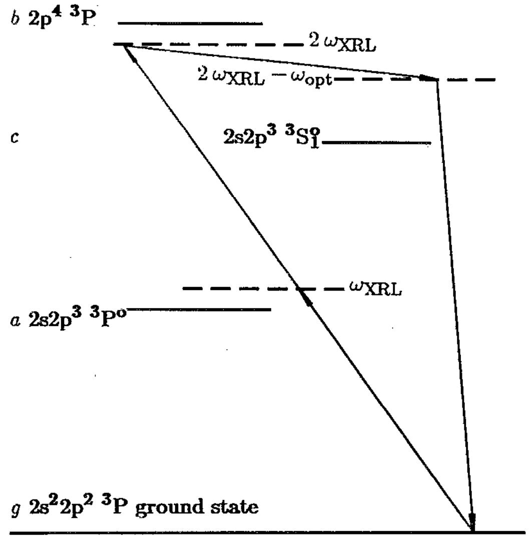

FIG. 1. C^{5+} x-ray laser four-wave mixing in C-like Ca XV.

$$C_{\text{eff}} \equiv P/P_1 = KN^2 P_1 P_2 |F_2|^2,$$
$$K \simeq 512\pi^6 c^{-2}(1-\lambda_1/2\lambda_2)\lambda_1^{-3}\lambda_2^{-1}(\Sigma\alpha S)^2. \quad (2)$$

Here P is the generated power; $N(\text{cm}^{-3})$ is the number density of the ions at the initial level g; $P_1(\lambda_1)$ and P_2 (λ_2) are the incident power (wavelength) of the XRL and the optical laser, respectively, in W/cm^2 (cm); $|F_2|^2$ is the phase-matching factor. The factor $(\Sigma\alpha S)^2$ reflects the energy-level structure of the ion as well as resonant conditions. Each S term in this sum corresponds to one of the contributing (near-resonant) combinations of the ion energy levels g, a, b, and c, and can be written as follows:

$$S^2 = A_{ag}A_{ba}A_{bc}A_{cg}[(E_a-E_g)(E_b-E_a) \times (E_b-E_c)(E_c-E_g)]^{-3} \times [(E_a-E_g-\lambda_1^{-1})(E_b-E_g-2\lambda_1^{-1}) \times (E_c-E_g-2\lambda_1^{-1}+\lambda_2^{-1})]^{-2}, \quad (3)$$

where E_i, $i=a,b,c,g$, is the energy of the corresponding level in cm^{-1}, and A_{ik} are the transition probabilities. Only the level g is assumed as populated substantially. Each coefficient α accounts for the angular momenta of the contributing states and for the signs of the dipole moments and of frequency detunings, and can be calculated using Refs. 6 and 7. Atomic information required for the calculation of S terms can be found in Ref. 8.

Our estimates of the coefficients K are listed in Table I. To calculate the conversion efficiency, the following output power of participating lasers is assumed: 10^6 W for Se^{24+} 206.38 and 209.78 Å lines and for the Y^{29+} 155.0 Å line; 10^5 W for the C^{5+} 182.097 Å line and the Ag^{37+} 99.36 Å line; 5×10^4 W for Ta^{45+} 44.83 Å line; 10^{12} W for the KrF 0.2484 μm laser; and 10^{12}, 10^{11}, and 10^{10} W for the Nd 1.06 μm laser fundamental, second, and fourth harmonic, respectively. (In most of the cases considered, an order of magnitude lower optical power may be enough for high conversion efficiency.)

The phase-matching factor $|F_2|^2$ is directly related to the dispersion. The refractive index due to free plasma electrons may be expressed as

$$n_f(\lambda) = 1-(2\pi)^{-1} r_e N_e \lambda^2, \quad (4)$$

where r_e is the classical electron radius and N_e is the plasma electron density. Refraction due to bound electrons is negligible compared to the free-electron refraction for all the media in Table I owing to the absence of close enough resonances between the incident and generated radiation and ionic transitions. As a result, the phase mismatch for the process (1) with *collinear* x-ray and optical beams can be estimated as follows:

$$\Delta k \simeq -r_e N_e \lambda_2. \quad (5)$$

In order to increase the phase-matching factor, the product $b\Delta k$ should be made as close to zero as possible.[5] For collinear beams with fixed confocal parameters, this can be done by adjusting the electron density. For example, for assumed $b=0.15$ cm and $\lambda_2 = 2.484\times10^{-5}$ cm (KrF laser), it follows from Eq. (5) that $b\Delta k \simeq -1$ for $N_e \simeq 10^{18}$ cm^{-3}. It yields $|F_2|^2 \simeq 4$,[5] which is only two times smaller than the optimal value of eight (for zero phase mismatch $\Delta k=0$). Optimal phase matching may be provided for *noncollinear* beams.[6] To do that, an XRL beam should be split into two beams. Then, phase mismatch is equal to zero if the angle between the optical beam and one of the x-ray beams is equal to $\theta \simeq [r_e(2\pi)^{-1} N_e \lambda_1 \lambda_2]^{1/2}$, whereas the angle between two x-ray beams is equal to $(2\pi - 2\theta)$.

For the sake of uniformity, all the values of C_{eff} listed in Table I are estimated for $N_e = 10^{18}$ cm^{-3} and $|F_2|^2 \simeq 4$.

TABLE I. Frequency upconversion of x-ray laser radiation in four-wave mixing. * denotes conversion efficiency approaching one.

Medium E_{ion}	Lasers	K	λ	C_{eff}
1. Ar VIII 143 eV	Ge^{22+} 232.24Å–Nd 1.06 μm	2×10^{-52}	117.41 Å	*
2. K IX 176 eV	Se^{24+} 209.78 Å–Nd 0.53 μm	10^{-52}	106.99 Å	*
3. Ca X 271 eV	Se^{24+} 206.38 Å–Nd 0.53 μm	5×10^{-51}	105.24 Å	*
4. K IX 176 eV	Se^{24+} 206.38 Å–Nd 0.266 μm	10^{-51}	107.35 Å	*
5. K IX 176 eV	C^{5+} 182.173 Å–Nd 0.263 μm	3×10^{-56}	94.29 Å	0.0002
6. Ca X 271 eV	Y^{29+} 155.0 Å–Nd 0.5320 μm	10^{-54}	78.65 Å	*
7. V XIII 336 eV	Ag^{37+} 99.36 Å–Nd 1.06 μm	10^{-53}	49.91 Å	*
8. Cu XIX 670 eV	Ta^{45+} 44.83 Å–Nd 1.06 μm	4×10^{-56}	22.46 Å	0.01
9. Ca XV 894 eV	C^{5+} 182.097 Å–KrF 0.2484 μm	10^{-53}	94.51 Å	*
10. Ar XIII 686 eV	Se^{24+} 209.78 Å–KrF 0.2484 μm	6×10^{-57}	109.51 Å	0.03

Our quantitative results should be viewed as only order-of-magnitude estimates, largely because at this stage we neglect all the competing processes. (It is worth noting, however, that substantial photoabsorption is unlikely for all the cases considered, due to relatively low density and the small length of plasma, and due to the absence of close resonances.) Nonetheless, these estimates allow one to expect substantial output power at almost doubled frequency for a number of available XRL's.

Tunable x-ray radiation may result from four-wave mixing (1) with tunable optical lasers. In particular, a tunable laser with the wavelength near 5000 Å and the power P_2 would produce broadly tunable output $P \simeq 10^{-5} \times P_2$ at almost doubled XRL frequency by mixing with C^{5+} XRL in Ca XV plasma or with Se^{24+} 209.78 Å XRL in Ar XIII plasma (from lines 9 and 10 of Table I, respectively). For P_2 of a few MW, Eq. (2) yield converted power of tens of watts which may be enough for the purposes of x-ray linear spectroscopy.

In conclusion, we have identified and evaluated resonant nonlinear media (plasmas) for efficient frequency up-conversion of a number of the available x-ray lasers by difference-frequency mixing with optical lasers. Substantial generation of tunable x-ray radiation is also expected in some of these media. Future work will include further analysis and optimization of this and other schemes, followed by design of a proof-of-principle experiment.

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