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Discharge-plasma – x-ray-laser resonant couples for x-ray nonlinear optics

P. L. Shkolnikov and A. E. Kaplan
Department of Electrical and Computer Engineering, The Johns Hopkins University, Baltimore, Maryland 21218
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We demonstrate the feasibility of x-ray resonant nonlinear effects (absorption saturation and nonlinear refractive index) for the radiation of some of existing x-ray lasers (XRL's) when it propagates in relatively low-temperature discharge plasmas different from those used in the respective XRL. We identified 12 resonant "XRL-plasma" couples suitable for such effects.

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We have recently demonstrated [1] the feasibility of saturation-related third-order x-ray resonant nonlinear effects (XRNE's) in x-ray-laser (XRL) and laserlike plasmas. The choice of the XRL ions as a medium for this XRL radiation nonlinear interaction has been justified by the perfect frequency match between the XRL radiation and the transition of interest. In such a case, however, we have to deal with very hot plasma (electron temperature $T_e > 1000$ eV). Moreover, the nonlinear transition coincides with the lasing transition, i.e., occurs between two *excited* levels, which are essentially empty for relatively low density of XRL-like plasma assumed in Ref. [1]. In order to make a nonlinear interaction with the XRL radiation observable, one has to have an additional x-ray radiation (pumping) resonant to the transition between the ion ground level and the lower laser level. This is required to significantly populate the latter by transferring ions from the ion ground level, which makes the proposed scheme most suitable for observation of the nonlinear optical effects inside the XRL itself.

To study the nonlinear optics interaction of XRL radiation with non-XRL materials, in particular plasmas, it would be much more desirable to have much *cooler* (i.e., less ionized) plasmas whose ions could also be excited by direct transition from their ground levels. Thus, the main purpose of this paper is to identify, as media for XRNE's the non-XRL ions, for which (i) the degree of ionization (and therefore, the required temperature) is significantly lower than that of the XRL plasma, so that such plasmas could be created much easier (preferably, by standard discharge devices) and (ii) the XRL radiation is matched to some direct transition from those ions' *ground* level so that no additional x-ray pumping would be required for the XRNE's observation. In this paper, we identify 12 such resonant couples "XRL line—plasma transition from the ground level" and demonstrate the feasibility of saturation-related XRNE's in five of them. Required moderate electron temperature and density (up to 350 eV and up to 10^{18} cm^{-3}, respectively) of plasma are readily attainable in the existing discharge devices.

A plasma is a good candidate for XRNE observation if the resonant detuning between the XRL frequency ν and the frequency of some transition from the ground to excited levels of the plasma ions ν_0 is less than the Doppler full width, $\Delta\nu^D$, on half maximum of the respective plasma transition. Several couples found by us are presented in Table I (in which the asterisk denotes wavelengths taken from Ref. [2] for the corresponding XRL transitions). Search for such resonances is complicated by the fact that too few accurately measured or calculated wavelengths of ion transitions (especially those of highly ionized atoms) can be found in the literature. Furthermore,

TABLE I. Resonant couples "XRL radiation–plasma-ion transition." A denotes the corresponding transition probability, χ denotes the plasma ionization potential, λ and λ_0 denote the wavelengths.

	XRL λ (Å)	Plasma λ_0(Å)	A (sc^{-1})	χ (eV)
1	Ge^{22+} 236.26	Ar *XIII* 236.27	3×10^9	686
2	Se^{24+} 220.28	Sc *IV* 220.280	6×10^8	73
3	Se^{24+} 209.78	Fe *X* 209.776	1.3×10^8	262
4	Se^{24+} 209.78	Cl *XIII* 209.81	10^{10}	656
5	Ge^{22+} 196.06	Fe *VII* 196.046	4×10^{10}	125
6	C^{5+} 192.097*	Na *IV* 182.123	1.4×10^{10}	99
7	Al^{10+} 105.69*	Na *IV* 105.6867	2.4×10^{10}	99
8	Al^{10+} 105.69*	Cr *VIII* 105.69	4.3×10^{10}	185
9	O^{7+} 102.355*	Ni *IX* 102.340	1.9×10^{11}	193
10	Eu^{35+} 71.00	Mg *VIII* 71.007		266
11	Ta^{45+} 44.83	Si *X* 44.83		401
12	W^{46+} 43.18	Cl *IX* 43.168	5×10^{11}	400

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oscillator and transition strengths are often unavailable, particularly for transitions from inner shells or transitions to highly excited states, which is often the case. Although this makes the list of resonant couples in Table I still open for new candidates, this table reveals significant opportunities.

Since, at this point, our main goal is to identify as many couples as possible (which is essentially fulfilled by the content of Table I), we will make an estimate of nonlinearity only for couples with special features: (i) couples 1 and 12 having the longest and shortest wavelength in the list, respectively, (ii) couple 4 involving the radiation of the most successful XRL so far, and (iii) couple 7 featuring plasma with one of the lowest ionization potentials in the list. The treatment similar to that for the above-mentioned couples can be applied to the rest of Table I. (Couple 2 will also be considered by us elsewhere in connection to a vuv laser with XRL resonant pumping.) Note that couples 10 and 11 have been included because of very good resonance, although we have not found the respective transition probabilities A in the literature.

In our evaluation of each couple we follow the same procedure. We first solve the system of rate equations for the population at the levels of interest in the steady-state approximation. This is quite adequate for couples 1, 4, and 12, for which the lifetime of the upper levels is shorter than the XRL pulse duration $\simeq 10^{-10}$ sec. For couple 7, this approximation is less adequate, but this can hardly affect the qualitative results. Each plasma temperature is chosen at half of the ionization potential; such a choice usually assures a large enough fraction of the ions of interest in the plasma. For the assumed plasma density $N_e = 3\times 10^{17}$ cm^{-3}, collision rates are essentially negligible compared to radiative rates, except for transitions between levels of the fine structure. As a result, many levels can be excluded from the calculations. At the same time, such a density appears to be high enough to produce significant XRNE's. Following a standard approach [3], only homogeneous broadening is taken into account at this step.

Solving the respective system of rate equations for the population density difference $\Delta N \equiv N_g/g_g - N_u/g_u$, where $N_g(g_g)$ and $N_u(g_u)$ are the population densities (statistical weights) of the ground level and the upper level of the transition of interest, respectively, we find that, for all the couples in consideration, ΔN can be written in the same form as for a two-level system:

$$\Delta N = \Delta N_0 \left[1 - \left[\frac{\Delta\nu}{2} \right]^2 \frac{I}{I^s} \frac{1}{(\nu-\nu_0)^2 + (\delta\nu)^2} \right] , \tag{1}$$

where $\Delta\nu$ is the homogeneous full width at half maximum of the transition, I is the intensity of the incident XRL radiation, $\delta\nu = (\Delta\nu/2)\sqrt{1+I/I^s}$, $\Delta N_0 = \Delta N(I=0)$, and that the "saturation intensity" I^s is as

$$I^s = I_0^s R , \quad I_0^s \simeq 785\lambda^{-3}\Delta\nu \quad (R<1) \tag{2}$$

(with λ in Å, I and I^s in W/cm^2). Here I_0^s corresponds to the saturation intensity of the two-level system with the statistical weights of all the levels not taken into account. The constant R in Eq. (2) can be regarded as the effective (dimensionless) relaxation parameter of the process. If the intermediate levels delay the relaxation only slightly, then $R \simeq 1$; it decreases when more ions accumulate at the intermediate levels. The specific expressions for R include the oscillator and collision strengths as well as the statistical weights of all the participating levels and vary from one couple to another. For example, for couple 4, only one intermediate level can be taken into account, in which case $R = X\tau(g_1 g_3^{-1} + g_2 g_1^{-1})$. Here X is the rate of collision excitation from the ground level to the intermediate level, τ is the lifetime of the upper (resonant) level, and g_1, g_2 and g_3 are the statistical weights of the ground, intermediate, and upper levels, respectively.

At this point we introduce inhomogeneous broadening by integrating Eq. (1) over the Doppler distribution [3], which determines the absorption coefficient $\gamma(\nu)$ and nonlinear correction $\Delta n^{\mathrm{NL}}(r) = n(r) - n(r=0)$ to the refractive index $n(r)$ in the presence of homogeneous or/and inhomogeneous Doppler broadening:

$$\gamma(\nu) = \gamma_0 \frac{\mathrm{Re}\, w(x+ib)}{\sqrt{1+r}} , \quad \gamma_0 = \frac{\sqrt{\pi \ln 2}}{4\pi^2} \frac{\lambda^2 A}{\Delta\nu^D} \frac{g_u}{g_l} N_i , \tag{3}$$

$$\Delta n^{\mathrm{NL}} = (4\pi)^{-1}\lambda\gamma_0[\mathrm{Im}\, w(x+ib) - \mathrm{Im}\, w(x+ib_0)] , \quad b_0 = b(r=0) . \tag{4}$$

Here γ_0 is the small-signal absorption coefficient at the central wavelength of the transition, $b = \sqrt{\ln 2}(\Delta\nu/\Delta\nu^D)\sqrt{1+r}$ describes the degree of homogeneous broadening, $r = I/I^s$ and $x = 2\sqrt{\ln 2}(\nu_0 - \nu)/\Delta\nu^D$ and the dimensionless intensity and detuning of the XRL radiation, respectively, and $w(x+ib)$ denotes the complex error function (see, e.g., Ref. [4]). The ion density N_i can be obtained [5] from the electron density N_e by dividing the latter by the fractional abundance of the ionization stage of interest and by the average ion charge. We assume $N_i \simeq 0.03 N_e$ for couples 1, 5, and 12, and $N_i \simeq 0.04 N_e$ for couple 7.

In our calculations we assume that the homogeneous linewidth is determined mainly by the lifetime of the upper level, $\Delta\nu \simeq A/2\pi$. As a result, even for rather moderate values of T_e, the homogeneous linewidth is significantly smaller than the Doppler linewidth. However, this "Doppler predominance" can easily be inhibited by even very modest XRL intensities so that use of full formulas, Eqs. (3) and (4), may be required.

For the data of Table I and frequencies therein, Eqs. (3) and (4) yield results summarized in Table II for the four couples. (For the estimates of the XRL intensities, we use data for the existing systems [6,7] in the case of couples 1 and 4, and some reasonable values for couples 7 and 12). For these intensities ($I \gg I^s$), the plasmas are essentially transparent because of the absorption saturation [$\gamma(\nu) \simeq 0$]. The last column in Table II contains the estimates of the length $L = \lambda/\Delta n^{\mathrm{NL}}$ at which the significant change ($\simeq 2\pi$ across the beam) could appear as

TABLE II. Saturation-related XRNE's in plasmas.

Couple	Sources of data	I (W/cm^2)	I^s (W/cm^2)	Δn^{NL}	L (cm)
1	[2],[8],[9]	10^9	600	3×10^{-7}	6
4	[2],[10]	2×10^9	5×10^3	10^{-7}	18
7	[2],[11],[12]	10^{10}	2×10^5	7×10^{-8}	16
12	[2],[13]	10^{12}	8×10^8	2×10^{-8}	20

a result of the nonlinear refractive index for given intensity I.

One can readily see from Table II that XRNE's resulting from nonlinear refractive index can be observed with either existing XRL intensities or those to be soon available. The expected effects based on the nonlinear refractive index are self-focusing and "running foci" [14] (although, because of the significant probability of multiphoton processes, we do not expect a pronounced wave collapse, in contrast with the situation in liquids and solids), self-trapping of XRL radiation similar to the one observed in Na vapor [15], soft-bending [16] (also observed in Na vapor [17]), and four-wave mixing [18]. Any of these effects may also be instrumental in the direct measurement of the nonlinear refractive index. Aside from XRNE's related to the nonlinear refractive index, other coherent effects such as 2π solitons and self-induced transparency [19] are also feasible. The experimental observation of XRNE's in resonant couples of Table I is greatly facilitated by the fact that the required parameters T_e, N_e, and L of plasmas can be attained by using standard discharge devices (e.g., modified z pinch [20], gas-linear pinch [21], and θ pinch [22]).

In conclusion, we identified 12 resonant couples, "XRL-discharge plasmas," suitable for the observation of nonlinear x-ray-laser propagation effects in relatively low-ionized discharge plasmas. Our estimates showed that these nonlinear effects are observable by using currently available XRL intensities and plasma discharge devices.

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[1] P. L. Shkolnikov and A. E. Kaplan, Opt. Lett. **16**, 1153 (1991).

[2] R. Kelly, *Atomic and Ionic Spectra Below 2000 Angstroms: Hydrogen through Krypton,* Natl. Bur. Stand. (American Chemical Society, New York, 1987).

[3] A. Corney, *Atomic and Laser Spectroscopy* (Clarendon, Oxford, 1977), Chap. 13; A. Yariv, *Quantum Electronics* (Wiley, New York, 1989), Chap. 8.

[4] *Handbook of Mathematical Functions,* Natl. Bur. Stand. Appl. Math. Ser. No. 55, edited by M. Abramowitz and I. A. Stegun (U.S. GPO, Washington, DC, 1964), pp. 297–304 and 325–328.

[5] B. L. Whitten, R. A. London, and R. S. Walling, J. Opt. Soc. Am. B **5**, 2537 (1989).

[6] C. J. Keane, N. M. Ceglio, B. J. MacGowan, D. L. Matthews, D. G. Nilson, J. E. Trebes, and D. A. Whelan, J. Phys. B **22**, 3343 (1989).

[7] H. Daido (private communication).

[8] A. K. Bhatia, J. F. Seely, and U. Feldman, At. Data Nucl. Data Tables **36**, 453 (1987).

[9] B. C. Fawcett, At. Data Nucl. Data Tables **37**, 367 (1987).

[10] A. K. Bhatia, U. Feldman, and J. F. Seely, At. Data Nucl. Data Tables **35**, 319 (1986).

[11] A. K. Bhatia, U. Feldman, and G. A. Doscheck, Astron. Astrophys. **80**, 22 (1979).

[12] B. C. Fawcett, At. Data Nucl. Data Tables **34**, 215 (1986).

[13] B. C. Fawcett, At. Data Nucl. Data Tables **31**, 485 (1984).

[14] Y. R. Shen, Prog. Quantum Electron. **4**, 1 (1975); J. H. Marburger, *ibid.* **4**, 35 (1975).

[15] J. E. Biorkholm and A. Ashkin, Phys. Rev. Lett. **32**, 129 (1974).

[16] A. E. Kaplan, Pis'ma Zh. Eksp. Teor. Fiz. **9**, 58 (1969) [JETP Lett. **9**, 33 (1969)].

[17] G. A. Swarzlander, Jr., H. Yin, and A. E. Kaplan, Opt. Lett. **13**, 1011 (1988); J. Opt. Soc. Am. B **6**, 1299 (1989).

[18] R. W. Hellwarth, Prog. Quantum Electron. **5**, 1 (1977).

[19] S. L. McCall and E. L. Hahn, Phys. Rev. Lett. **18**, 908 (1967); Phys. Rev. **183**, 457 (1969).

[20] R. Noll, H. Kinze, and C. R. Haas, Nucl. Instrum. Methods Phys. Res. A **278**, 85 (1989).

[21] K. H. Finken, in *Spectral Line Shapes,* edited by K. Burnett (de Gruyter, New York, 1983), Vol. 2, p. 117.

[22] R. C. Elton, R. V. Datla, J. R. Roberts, and A. K. Bhatia, Phys. Rev. A **40**, 4142 (1989).