Feasibility of x-ray resonant nonlinear effects in plasmas

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Received March 18, 1991

We demonstrate the feasibility of saturation-related third-order x-ray resonant nonlinear effects, in particular, absorption saturation and nonlinear refractive index in x-ray laser and laserlike Se XXV and Mo XXXIII plasmas as well as in other plasmas with lower degrees of ionization.

Recent developments in x-ray laser (XRL) research have resulted in the experimental observation of laser amplification at many wavelengths in the soft-x-ray domain,[1] ranging from $\lambda = 28$ nm to $\lambda = 4$ nm. Within the next few years powerful sources of coherent XRL radiation at those wavelengths will also be available. This sets the stage for research on the interaction of intense coherent x-ray radiation with matter, in particular, on x-ray nonlinear optics.

The first obvious choice of the environment for the x-ray resonant nonlinear effects (XRNE's) to occur and to be experimentally observed in is the plasma consisting of the same ions that give rise to the laser action itself. XRNE's in these situations are expected to be essentially similar to nonlinear effects due to other resonantly enhanced nonlinear interaction of light with matter in a visible optical domain. In this Letter we present an evaluation of the saturation intensity and nonlinear refractive index for the soft x rays in the Se XXV and Mo XXXIII x-ray laser and laserlike plasmas. We also briefly discuss the feasible nonlinearity of nonlaser plasmas with lower degrees of ionization, such as Fe X, Na IV, and Cl XIII, whose frequencies closely match those of XRL's. The investigation of the former group of plasmas is important since their XRNE's transitions are essentially the same as the lasing transitions of their respective XRL's. For the latter group of plasmas, a good resonance between some of the transitions from the ground level and XRL radiation, as well as the relatively low temperature of those plasmas, may provide good conditions for the XRNE's observation.

The strongest nonlinear x-ray interactions may occur in the active XRL plasma, e.g., in the neonlike Se or Mo plasma, during x-ray lasing. However, from the point of view of studying XRNE's as such, i.e., in a situation in which they do not intervene with the lasing process, it would be more instructive first to investigate plasma that consists of the same ions as the respective XRL but with an electron density N_e not high enough to excite laser action. This assumption also significantly simplifies[2] the problem. In particular, it allows us to neglect collision rates for the transitions near the neonlike ground state and to take into account only the levels coupled by x-ray radiation of interest. At the same time the stability of neonlike plasmas in a wide range of N_e ensures that the fraction of the neonlike ions f_{Ne} in the plasma will remain almost unchanged. In the absence of any external pumping radiation between the ground level and the $3p$–$3s$ levels, such low electron density cannot provide any significant $3s$–$3p$ level population (see, e.g., Ref. 3 for Kr), and therefore no interaction with the XRL radiation resonant to $3s$–$3p$ transitions can be observed. However, XRNE's can be made feasible if the $3s$ level is populated, e.g., by the powerful radiation resonant to the transition between the $3s$ and the ground levels. This radiation conveniently originates from the radiative decay of the $3s$ level in the XRL.

As a result, in the case of neonlike plasmas, our model could be restricted to only three levels: the upper (u) and the lower (l) XRL levels and the neonlike ground level (g). Levels u and l in this low-density plasma will be coupled by the XRL radiation with the resonant frequency; we are interested in the absorption saturation and nonlinear refraction at that transition. Level l will be populated by sufficiently strong (incoherent) pumping radiated as a side product by the respective XRL as a result of the decay of its level l into its ground level g. Two examples will be considered, Se XXV as the medium of the most successful XRL so far[1,4] and Mo XXXIII, for which gain for the x-ray with one of the shortest wavelengths in the neonlike sequence has been recently reported.[4] Energy-level diagrams simplified for our purpose for the neonlike Se and Mo ions are given in Fig. 1.

The first important problem to be solved is the choice of the electron temperature T_e. On one hand, one should try to choose the electron temperature T_e as low as possible to decrease the Doppler broadening. On the other hand, it appears to be impossible to make use of the plasma with $T_e \ll 1/2\chi$, with χ being the ionization potential of the neonlike ion, since in such a case the fraction of neonlike ions f_{Ne} would be too small,[5] and this drawback cannot be compensated for by the Doppler broadening's decreasing. Hence, to have a meaningful estimate, we choose $T_e = 1/2\chi$, i.e., T_e equal to the temperature of the lasing plasmas.[5–7] We also assume an ion temperature of $T_i = 0.4T_e$, as in Ref. 5. (This means

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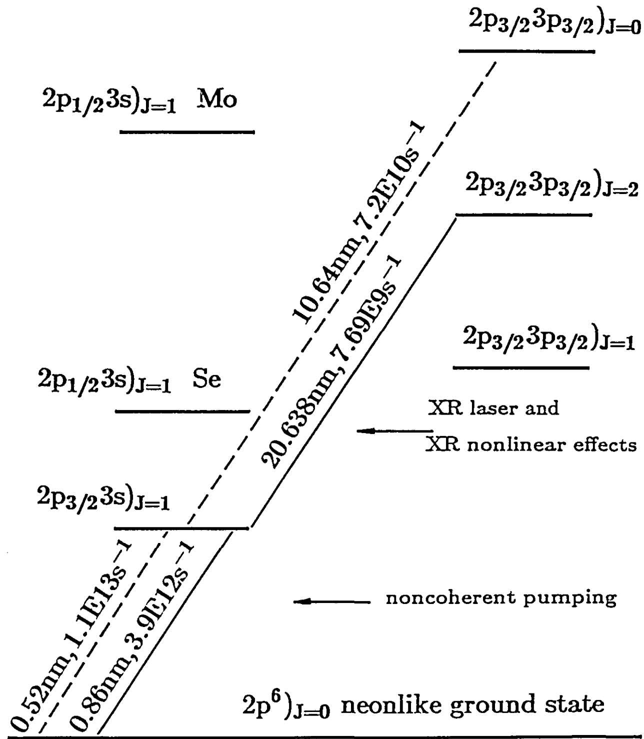

Fig. 1. Simplified energy-level diagram for neonlike ions. Wavelengths and spontaneous emission rates are shown for transitions of interest in Se XXV (solid lines) and Mo XXXIII (dashed lines). Note that the level $2p_{1/2}3s)_{J=1}$ is at different locations for the Mo and Se ions.

that $T_i \simeq 400$ eV for Se and 800 eV for Mo.) For the assumed electron temperatures, f_{Ne} is almost constant for a wide range of N_e below the lasing optima and may be assumed to be 0.3 and 0.1 for Se and Mo, respectively.[5] Dividing f_{Ne} by the average ion charge, one obtains the ratio α of neonlike ion density of N_i to N_e equal to 0.02 for Se and to 0.005 for Mo. Collision rates can be neglected if $N_e \ll A_{ik}/C_{ik}$, where A_{ik} and C_{ik} are the radiative and downward collisional rate coefficients for transitions of interest,[2] respectively. The atomic data from Refs. 5–8 and the principle of detailed balance (required to obtain downward collisional rate coefficients from upward ones[9]) yield $N_e \simeq 10^{18}$ cm^{-3} for Se plasma and $N_e \simeq 10^{19}$ cm^{-3} for Mo plasma.

Since the levels u and g are not coupled by radiative processes (in the dipole approximation), we can break our three-level system into two two-level subsystems consisting of the levels l–g and u–l, respectively. As the first step, we determine the pumping intensity I_{lg} for the radiation with the central wavelength λ_{lg} of the l–g transition, necessary to produce a steady-state population density in the level l, sufficient for observation of nonlinear effects at the u–l transition. Then we consider the nonlinear behavior of the absorption coefficient and refractive index for the XRL radiation resonant or near resonant to the u–l transition, assuming that the total sum of populations at the l and u levels depends only on the pumping and not on the XRL radiation intensity. The XRNE estimates based on this approximation may differ by a factor of less than 2 from those for a full three-level model. Two-level models[10,11] modified to take into account the level degeneracy[12] and the inhomogeneous broadening,[13] result in the following set of equations. At first, for the dimensionless l level population in the absence of the XRL radiation $\beta \equiv N_l/N_i$ (N_l denotes the population density of the l level), we have

$$\beta = \sqrt{\pi \ln 2}\,(1 + g_l^{-1})^{-1} \Delta_{lg} r_{lg} (1 + r_{lg})^{-1/2}, \quad (1)$$

where Δ_{lg} is the ratio of the Lorentzian FWHM $\Delta\nu_{lg}$ to the Doppler width $\Delta\nu_{lg}^D \equiv 2\nu_{lg}[(2kT_i/M_i c^2)\ln 2]^{1/2}$ for the l–g transition, ν_{lg} being the central frequency, M_i is the mass of the ion, $r_{lg} \equiv I_{lg}/I_{lg}^s$ is the dimensionless pumping intensity with $I_{lg}^s = 4\pi^2 hc\,\Delta\nu_{lg}\lambda_{lg}^{-3}(1 + g_l)^{-1}$ being the l–g saturation intensity, and g_l is the statistical weight of the level l. This equation is valid unless r_{lg} is large enough (>50) to suppress the implied Doppler-broadening predominance. It is worth noting that no significant absorption of the pumping photons with the energy $h\nu_{lg}$ by the excited neonlike ions is expected, since the energy $2h\nu_{lg}$ is significantly higher than the ionization potential of these ions.

For the absorption coefficient $\gamma(\nu)$ of the incident radiation with the frequency ν, one obtains

$$\gamma(\nu) = \gamma_0 \frac{\mathrm{Re}\, w(x + ib)}{\sqrt{1 + r}},$$

$$\gamma_0 \equiv \frac{\sqrt{\pi \ln 2}}{4\pi^2} \frac{\lambda_{ul}^2 A_{ul}}{\Delta\nu_{ul}^D} \frac{g_u}{g_l} \beta\alpha N_e, \quad (2)$$

where λ_0 is the small-signal absorption coefficient at the central wavelength λ_{u-l} of the u–l transition, $b \equiv \sqrt{\ln 2}\,\Delta_{ul}\sqrt{1 + r}$ describes the degree of homogeneous broadening, $r \equiv I/I_{ul}^s$ and $x = 2\sqrt{\ln 2}\,(\nu_{ul} - \nu)/\Delta\nu_{ul}^D$ are the dimensionless intensity and detuning of the XRL radiation, respectively, $I_{ul}^s = 4\pi^2 hc\,\Delta\nu_{ul}\lambda_{ul}^{-3}$ is the saturation intensity, $w(x + ib)$ denotes the complex error function (see, e.g., Ref. 13), and g_u is the u–l level statistical weight. Finally, the nonlinear correction $\Delta n^{\mathrm{NL}}(r) = n(r) - n\,(r = 0)$ to the refractive index $n(r)$ can be written as

$$\Delta n^{\mathrm{NL}} = (4\pi)^{-1}\lambda_{ul}\gamma_0[\mathrm{Im}\, w(x + ib) - \mathrm{Im}\, w(x + ib_0)],$$

$$b_0 \equiv b(r = 0) = \sqrt{\ln 2}\,\Delta_{ul}. \quad (3)$$

In our calculations we assume that the homogeneous linewidths are determined mainly by the lifetime of the l–g transitions (similarly to those in Ref. 14), so that $\Delta\nu_{ul} = \Delta\nu_{lg} \simeq A_{lg}/2\pi$. Given the transitions data and plasma conditions, one can obtain the following estimations for Se (Mo) plasma. A pumping intensity $I_p \simeq 10^{13}$ (10^{14}) W/cm^2 is required to attain $\beta = 0.1$. Such a population yields the absorption coefficient at the central u–l frequency $\gamma(\nu_{ul}) \simeq 1.5$ (0.5) cm^{-1}, with the saturation intensity $I_{ul}^s \simeq 6 \times 10^7$ (10^9) W/cm^2. The nonlinear part of the refractive index is $\Delta n^{\mathrm{NL}} \simeq 3 \times 10^{-8}$ (10^{-8}) for $I = 3 \times I_{ul}^s$ and dimensionless detuning $x = 0.8$ (such a detuning corresponds to the frequency ν being within the kernel of the Doppler line shape).

Our calculations of nonlinearities in the laser active medium show that at least for Se XRL they should be large; this conclusion does not depend on any specific model. Indeed, the power of the order of 1 MW per line and the source size of $\simeq 200\ \mu m$ (Ref. 4) mean that the intensity $I \simeq 2 \times 10^9\ W/cm^2 \simeq 35 I_{ul}^s$ has been reached in the Se plasma. According to Eq. (1), this decreases the small-signal gain by a factor up to 20. [This result remains almost the same even if, in addition to the lifetime of the l–g transition as above, the collision dephasing time $\simeq 4 \times 10^{-13}$ s (Ref. 15) is taken into account.] Therefore the discrepancy between the calculated (38 cm^{-1}) and measured (5 cm^{-1}) small-signal gain coefficients[5] could be attributed at least in part to the gain saturation. Furthermore, even assuming the measured small-signal gain coefficient for γ_0, one obtains from Eq. (3) that for the intensities existing inside the Se XRL, the nonlinear correction to the refractive index can become fairly large in the x-ray domain, $\Delta n^{NL} \simeq 8 \times 10^{-8}$ to 2×10^{-7}. This nonlinear refractive index may result in the significant change ($\simeq 2\pi$ across the beam) in the phase front of the wave at the length $L_{NL} = \lambda/\Delta n^{NL} \simeq$ 5–10 cm.

Our choice here of the same ions as in the XRL plasma has been justified by the advantage of a perfect frequency match between the XRL radiation and the transition of interest. In such a case, however, we have to deal with a still hot plasma and to use additional x-ray irradiance (pumping), since the nonlinear transition occurs between two excited levels. It would be greatly desirable, therefore, to identify also some alternative (i.e., non-XRL) plasmas, for which (i) the degree of ionization (and therefore, the required temperature) is significantly lower than that of the XRL plasma and (ii) the XRL radiation is matched (not necessarily perfectly) to some direct transition from their ground level. More than 10 such XRL line-plasma couples have been found by us; some of the examples are the Se XXV 20.978-nm line and Fe X or Cl XIII plasma and the Al XI 15.07-nm or C VI 18.2-nm line and Na IV plasma. Our preliminary evaluations show that significant XRNE's can be feasible for $T_e \simeq 35$–300 eV and $N_e \simeq 10^{17}$–$10^{18}\ cm^{-3}$. Such plasmas can be readily produced by relatively simple and well-controlled electrical discharge.[16]

All these estimates indicate a significant potential for third-order XRNE's in XRL plasma and other resonantly absorbing plasmas. Self-action effects based on the resonant nonlinear effects such as self-(de)focusing, self-trapping, and self-bending can be observed rendering themselves as either desirable effects for enhancing of laser action or the effects to be avoided or inhibited. Other effects of the same group, e.g., four-wave mixing, may efficiently be used for plasma diagnostics and for phase-conjugation amplification. Since intensities significantly exceeding the saturation intensity have apparently been achieved in experiments, it may also be possible that self-transparency and related 2π solitons can be attained in a manner similar to that in the optical domain.[17] As a next step, high-order harmonics generation in the x-ray domain can be a natural extension of the similar optical effect. Our future plans include investigation of XRNE's in metallic vapors and, what may be the most promising, in condensed matter.

This research is supported by the U.S. Air Force Office of Scientific Research.

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