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Review of X-ray nonlinear optics

P. L. Shkolnikov and A. E. Kaplan

Electrical and Computer Engineering Department, The Johns Hopkins University, Baltimore, MD 21218

Abstract. We review the emerging sub-field of nonlinear optics -- nonlinear optics of X-rays. After briefly addressing the early theoretical and experimental work on the subject, we review in detail the recent theoretical research on soft-X-ray nonlinear optics. This research has already shown the feasibility of numerous resonant nonlinear optical processes (nonlinear absorption and refraction, third-harmonic generation, four-wave mixing, and stimulated Raman scattering) in plasma and gases irradiated by soft-X-ray lasers. Plasma media with ionic transition frequencies matching existing X-ray laser lines have been found and evaluated for potentially efficient transformations of the radiation of all the most powerful X-ray lasers reported to date. For X-ray stimulated Raman scattering, resonant non-ionized media (atomic gases and vapors) have been proposed and evaluated.

1. Introduction

Nonlinear optics was born in 1961, with the observation of the second harmonic of the ruby laser [1]. Since then, a vast majority of research in this, now well-developed, research area has been conducted with visible, IR , or near-UV radiation. Much smaller effort has been devoted to shorter wavelengths, due to two fundamental reasons: *(i)* all optical nonlinearities drop drastically as pumping frequencies increase, and *(ii)* short-wavelength lasing is much harder to attain. At the same time, nonlinear optics at short wavelengths is of substantial interest. On one hand, one may expect novel nonlinear effects or novel features of already known processes at very high frequencies capable of exciting inner-shell electronic or even some nuclear transition. On the other hand, short-wavelength nonlinear optics could find applications in such important areas as nonlinear spectroscopy of solids and plasma, and generation of even shorter-wavelength coherent radiation.

The first papers on X-ray nonlinear optics (XRNO) were published in the late 1960th -- early 1970th, following the first proposals for X-ray lasing [2]. In 1969, I. Freund and B. F. Levine [3,4] (see also [5]) showed theoretically feasibility of parametric conversion of X-rays in solids,

$$\omega_p \rightarrow \omega_s + \omega_i \qquad (1)$$

where ω_p, ω_s, and ω_i are the pump (X-ray), signal, and idle frequency, respectively. The source of nonlinearity for this process, with the energy of incident photons assumed to be much larger than the K-shell binding energy, is the dependence of the electron velocity in the Lorentz force on the applied electrical field [6]. The crystal structure of a solid medium provided for phase matching. In the unique pioneering experiments, P. Eisenberger and S. L. McCall [6], using the input flux of only 2×10^7 ph/s of 17 keV photons from an X-ray tube, managed not only to demonstrate the decay of a 17 keV photon into a pair of ≈ 8.5 keV photons, but also to show a quantitative agreement with the theory.

This early XRNO was developed almost exclusively in theory and largely by effort of its pioneers I. Freund, B. F. Levine, P. Eisenberger, and S. L. McCall. The subjects included: polarization properties of the X-ray parametric decay [4]; sum- and difference-frequency generation by coherent optical and X-ray radiation [7]; two-photon (X-ray + optical) absorption [8] (see also [9]); two-photon emission of X-rays [10]; oscillator models for

XRNO [11]; the nonlinear atomic scattering factor beyond the free-electron assumption [12]; feasibility of phase-matched three-wave mixing in crystals [13]; and the proposal to apply the process (1), with idle in EUV, to determining valence electron charge distribution in a crystal [14]. In the context of this last proposal, the second (and the last until the recent years) XRNO experiment was conducted [15].

Two distinct features characterized this early work. First of all, the lowest (second)-order X-ray nonlinear processes were considered (see also more recent Ref. [16]), apparently due to the absence of intense coherent X-ray sources at the time. Secondly, the photon energy assumed to be much higher than the electron binding energy precluded resonant enhancement; in fact, a resonance would decrease the efficiency of the parametric down-conversion [12]. The low available X-ray intensity and the lack of resonant enhancement predetermined the extremely low output of all the effects considered. As a result, although that early work has had some continuation [17], including experiments, the entire field of XRNO had to wait for the X-ray laser (XRL) to come.

The first X-ray amplification was observed in 1984 [18] at wavelengths close to 200 Å. Since then, XRLs have come a long way. Gain at ~*50* lines between 326 Å and 35 Å has been reported, with the output of Ne-like Se, Ge, and Zn XRLs reaching *3–10 MW*. Moreover, recently the output of *>30 MW* has been reported for Y XRL at LLNL, with the potential for the focused intensity of ~10^{18} *W/cm*^{2}. Ge XRLs are almost diffraction-limited, with polarized output, cavity operation, and oscillator-amplifyers schemes demonstrated.

This rapid progress renewed the interest in X-ray nonlinear optics: more than twenty publications have appeared since 1990 [19-39], with the first subjects being frequency transformations [19,20] and conceptually fundamental saturation-related X-ray resonant nonlinear effects. We began systematic theoretical research on XRNO with X-ray resonant nonlinearities inside an XRL itself [22,23], because they were important for X-ray lasing and because in this case we already had an exact resonance. Our results suggested that the X-ray field inside the active medium of XRLs that already existed at that time was more than sufficiently intense for substantial gain saturation. (In an earlier paper [21], gain saturation was included as part of a wave-optics model of a soft-X-ray laser, but no estimates of the magnitude of the effect was given.) Recent extensive calculations [39] arrived, in a much more sophisticated way, to the estimate of the gain decrease due to saturation close to the factor of twenty obtained in [23].

Our next step in search of feasible X-ray nonlinear effects was to move from the unique and hard to attain conditions inside of XRL active media to plasmas of much lower temperature and density, with soft-X-ray lasers in resonance with transitions from the ionic *ground* levels [24]. We have shown the feasibility of X-ray absorption saturation and nonlinear refraction for a number of existing X-ray lasers in discharge or laser-generated plasmas of moderate temperature and density.

Almost simultaneously, the feasibility of X-ray third-harmonic generation in plasma was theoretically demonstrated [25] in some plasmas of Li- and Na- like ions for available XRL lines (see also [27,28]). This effect, which is a method of choice for frequency upconversion of longer-wavelength radiation, is particularly important in X-ray domain where the pumping energy required to attain lasing at higher frequencies becomes prohibitively high very soon.

In the following, we review research on resonant soft-X-ray nonlinear optics. After addressing the properties of plasmas as X-ray nonlinear media (Section 2), we discuss the two topics of the largest experimental interest at this point: frequency transformations of soft-X-ray laser radiation by means of X-ray + optical four-wave mixing (Section 3, which also include some examples unpublished previously), and X-ray stimulated electronic Raman scattering (Section 4). Quite a few differences between Section 3 of this Review and the respective publications are largely due to more uniform plasma conditions adopted here.

2. Plasmas as X-ray nonlinear media

The advent of soft-X-ray lasers apparently removed one major obstacle on the road to X-ray (or, rather, soft-X-ray) nonlinear optics -- the lack of intense radiation sources; indeed, at optical wavelengths, the incident power in megawatts is more than sufficient for observing various nonlinear effects. With X-rays, however, observability of nonlinear effects requires also large resonant enhancement. Moreover, practically useful output of nonlinear frequency transformations, such as the third-harmonic generation, requires *multiple* resonant enhancement. From this point of view, plasmas are the first obvious candidates for X-ray nonlinear media, because there is a very larger variety of ions whose lines are in the X-ray domain. (Plasma media for short-wavelength nonlinear optics were first suggested by S. Harris back in 1973 [41].) In fact, although some non-ionized media could support XRNO effects (see Section 4 below), the majority of results in this field have been obtained for plasma. In this Section, we give general description of plasmas as X-ray nonlinear media.

In all the research on XRNO in plasma so far, it has been assumed that a homogeneous plasma is prepared *before* the X-ray radiation enters it; this precludes strong defocusing due to inhomogeneous electron distribution. Next, plasma electron density, N_e, is assumed much smaller than the critical density for all the participating wavelength, $N_{cr}(cm^{-3}) \approx 10^{21}\,\lambda^{-2}(\mu m)$; for instance, for $\lambda = 1\ \mu m$, $N_{cr} \sim 10^{21}\ cm^{-3}$, whereas for the X-rays at ~ 200 Å, $N_{cr} \sim 3\times 10^{24}\ cm^{-3}$. The roughly uniform electron density attainable on a routine basis is several orders of magnitude lower; we assume usually $N_e < 10^{19}\ cm^{-3}$. We also assume that the radiation of interest does not change the nonlinear medium, in particular does not ionize it further. As a result, plasma as an X-ray nonlinear medium may be seen as simply a collection of ions and electrons, such that ions are the sole source of optical nonlinearities, whereas free electrons determine the dispersion; bound electrons can substantially influence the plasma dispersion only for laser frequencies closer than ~3 Doppler width to an ionic transition frequency.

Further simplification comes from the stringent resonant requirements, which limit the dipole transitions to be accounted for in the nonlinear succeptibility to the few closely resonant ones. Our notion of resonance, though, is quite lax: since there is no tunable XRL yet, and the linewidth of the available XRLs reflects largely the Doppler width of ionic spontaneous emission ($\sim 10^{-4}$), we will call it "resonance" if the XRL detuning from an ionic transition is less than three Doppler width. In fact, except for the examples related to absorption saturation, it has always been possible to neglect resonant absorption, whereas the multiple resonant enhancement of the nonlinear effects of interest has been formidable.

Search for multiple-resonant plasma media is greatly complicated by the scarcity of accurate information on energy levels and especially on transition probabilities in ionized atoms. With the narrow selection of intense X-ray laser lines, this has made the search for resonant couples often more time-consuming than the calculations. Moreover, on several occasions, we have had to "borrow" the value of a transition oscillator strength from a neighbor in the isoelectronic sequence.

3. Four-wave mixing of coherent X-ray and optical radiation in plasma

Of all possible four-wave mixing (FWM) processes, the following one is most promising for efficient frequency transformations of XRL radiation:

$$\omega = \omega_{XRL1} + \omega_{XRL2} - \omega_{opt}\ ; \tag{2}$$

here ω_{XRL} is the frequency of an XRL (ω_{XRL1} and ω_{XRL2} may be the same or different), and ω_{opt} is the frequency of an optical laser. The main importance of this process lies in its ability to increase substantially the selection of available X-ray coherent sources, especially at shorter wavelengths. In spite of the rapid progress in X-ray laser research, the variety of coherent X-ray sources is still very limited. In the visible, IR, and UV domains, coherent radiation sources are greatly diversified by nonlinear optical transformations; such transformations may appear even more important in the X-ray domain. The conversion efficiency of the processes (2) is potentially very high, since *(i)* powerful optical lasers may be used; *(ii)* one of the participating frequencies is relatively low (optical); *(iii)* close resonances are more likely; and, finally, *(iv)* optimal phase matching is possible in a single-species plasma.

The first analyses of X-ray + optical difference-frequency mixing [19,26] were done in the loose-focusing geometry and did not target any particular XRL. In the subsequent publications [28-30,32,37], plasmas have been identified and evaluated in which efficient frequency transformations are expected for a number of available X-ray lasers by means of resonant phase-matched four-wave mixing with Nd or KrF lasers (or their harmonics or Stokes waves) in the tight-focusing geometry. If a *tunable* optical laser is used instead, the result would be tunable coherent X-ray radiation at almost doubled XRL frequency.

Only multiple-resonant four-wave mixing effects are feasible at the present level of X-ray laser technology; all the three resonances: one-photon, two-photon, and three-photon -- are required for high conversion efficiency, and at least two -- for modest efficient conversion. From this standpoint, Na-like and C-like ions appear to be most suitable for X-ray + optical four-wave mixing. For many C-like ions, the frequencies of the $2s^2 2p^2\ ^3P \rightarrow 2s2p^3\ ^3P^o$ and $2s2p^3\ ^3P^o \rightarrow 2p^4\ ^3P$ transitions are almost equal, which may provide one- and two-X-ray-photon resonances. Na-like ions may provide all three resonances for X-ray + optical FWM, with a disadvantage that initial levels of nonlinear transitions must be excited levels, so that additional effort might be necessary to populate these levels significantly. (Cu-like and Zn-like ions also have a potential for multiple resonances, but the lack of atomic information has not allowed so far to consider them as XRNO media [26].)

Our estimates of the conversion efficiency of X-ray + optical FWM are based on the general expressions for the power generated in the process (2) by Gaussian incident beams, with all frequencies different and all confocal parameters the same,*b* [see e. g. [42], Eqs. (40) and (41)]:

$$P = 576\pi^2 nc^{-2}(k_0^4 k_1 k_2 k_3 / k^2 k' n_1 n_2 n_3) N_i^2\ |\chi^{(3)}|^2 P_1 P_2 P_3\ |F_2|^2, \tag{3}$$

respectively. Here N_i is the number density of the ions in the initial level (the other resonant levels are assumed unpopulated); $\chi^{(3)}$ is the respective nonlinear succeptibility; $k_0 = \omega/c$; k is the wavevector of the generated radiation; $k_j, j = 1,2,3$ are the wavevectors of the incident radiation numbered by their places in the right-hand side of the Eq. (2); n, n_j are respective refractive indices; P_j are the incident radiation powers; and $k' \equiv k_1 + k_2 - k_3$. The term $|F_2(b\Delta k)|^2$ reflects phase-matching conditions. To take advantage of high intensity of focused beams, the tight-focusing is assumed, so that the ratio of the confocal parameter b to the plasma cell length L is small, $b/L \le 0.1$; in contrast to the third-harmonic generation, difference-frequency mixing can be optimally phase-matched for tightly focused beams in positively-dispersive media. The phase mismatch $\Delta k \equiv k - k'$ is determined almost totally by the wavelength of an optical laser,

$$\Delta k \approx -r_e N_e \lambda_3, \tag{4}$$

if (as we will always assume in this Section) free-electron dispersion dominates.

Introducing another phase-matching factor, G, to incorporate the entire explicit dependence of C_{eff} on the plasma density:

$$G = |b\Delta k F_2|^2\ , \tag{5}$$

and substituting conventional expressions for $\chi^{(3)}$ for only the resonant level combinations taken into account, we arrive to the following equation for the conversion efficiency $C_{eff} \equiv P/P_1$:

$$C_{eff} \approx 7.12\times 10^{-3}\ \lambda_3^{-2}(\lambda_1^{-2} + \lambda_1^{-1}\lambda_2^{-1})(\sum \beta S)^2 (N_i/N_e)^2 I_2 I_3 G \equiv K (N_i/N_e)^2 I_2 I_3. \tag{6}$$

In Eqs. (6), λ_i are in *cm*, and I_i are in W/cm^2. For $\omega_{XRL1} = \omega_{XRL2}$, the numerical factor in Eq. (6) is two times larger; such a process may be called "XRL frequency near-doubling".

The expression $(\sum \beta S)^2$ in Eq. (6) reflects the energy-level structure of the ion as well as resonant conditions, in the following way. Each S-term in this sum corresponds to one of the contributing (i. e. near-resonant) combinations of the ion energy levels g, a, b, and c:

$$S^2 = f_{ag} f_{ba} f_{ac} f_{cg} [(E_a - E_g)(E_b - E_a)(E_b - E_c)(E_c - E_g)]^{-3} \times \tag{7}$$
$$[(E_a - E_g - \lambda_1^{-1})(E_b - E_g - \lambda_1^{-1} - \lambda_2^{-1})(E_c - E_g - \lambda_1^{-1} - \lambda_2^{-1} + \lambda_3^{-1})]^{-2}\ ,$$

where E_i are the energy of (sub)levels in cm^{-1}, and f_{ik} are absorption oscillator strengths. Each β-term, of order of 0.2-0.5, is a product of four factors (one for a pair of the participating LS-sublevels), each equal to $\sqrt{(2l_l+)/l_>}\ T_{ik}$, where l_l is the orbital momentum of the lower sublevel, $l_>$ is the larger orbital momentum of the two. The angular matrix elements T_{ik} can be found in Refs. [43,44] (see also [26]).

The phase-matching factor, G, depends only on the geometry of the process (confocal parameters of the beams, their directions, location of the focal points, and the medium length), and of the medium dispersion. For collinear beams focused tightly to the same confocal parameter inside a medium, G is optimal, $G \approx 50$, if [42]

$$|b\Delta k|_{opt} \approx 2.7 \rightarrow G \approx 50; \tag{8}$$

the respective optimal electron density N_e^{opt} is as

$$N_e^{opt}\,(cm^{-3}) \approx 3.6\times10^{17}\ b^{-1}\ (cm) \tag{9}$$

Estimates of the coefficients K, Eq. (6), for the optimal phase matching, Eq. (8), are given in Table 1 for several resonant combinations of lasers and ions. With a reasonable ratio $N_i/N_e \sim 0.01$, terawatt optical lasers, and available XRL output, these estimates yield very high conversion efficiency, of up to a few per cent. Further increase is possible e. g. by increasing the ratio N_i/N_e or by tighter focusing.

With expected high efficiency of multiple-resonant difference-frequency mixing of X-ray and optical radiation, one could in principle attain much shorter wavelength using *cascades* of such mixings to provide a bridge between powerful Y, Se, and Ge XRLs and the "water window" [30].

Tunable coherent X-rays can also be generated by difference-frequency mixing of coherent X-ray and optical radiation. For instance, by mixing Se XRL 206 Å and 209 Å lines with the lowest harmonics of Nd or KrF lasers in K IX or Ca X, one may attain "line-by-line" tunability near 107 Å with high C_{eff} (possible output at 105.24 Å, 106.28 Å, 107.00 Å, 107.35 Å, 108.08 Å, 108.12 Å, and 112.54 Å). A tunable laser with the wavelength near 5000 Å and the power P_2 would produce a tunable output $P \approx 10^{-5}\times P_2$ 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 1, respectively).

4. X-ray stimulated electronic Raman scattering in non-ionized media

The vast majority of all the media proposed for X-ray resonant nonlinear optics have been plasmas. The feasibility of X-ray nonlinear effects in *non-ionized materials,* interesting theoretically and important experimentally (since it is much easier to work with neutrals), depends on whether resonances to XRL lines exist in neutral atoms, and whether the processes of interest would have time to develop before the medium becomes totally ionized by intense X-rays. We proposed [31,33,34], for the first time, two schemes for observing a resonant X-ray nonlinear effect, stimulated electronic Raman scattering (SERS), in non-ionized media -- Li vapor and He, and showed that substantial conversion efficiency could be achieved with high pumping energy capable of totally ionizing a gas within a time period much shorter than the pulse duration. We have studied in detail the dynamics of the process and predicted soliton-like pulses and precursors at the Stokes frequency at the photoionization front of pumping X-ray radiation [35]. Very recently, new calculations [40] have shown even more promise for the experimental realization of X-ray SERS.

X-ray SERS could be observed only if resonantly enhanced by tuning the pumping frequency close to some transition from the initial Raman level. Since X-ray laser photon energy (50-300 eV) is much higher than the binding energy of optical electrons in all the neutral atoms, we propose making use of so called core-exited (multiple-exited) atomic states, see e. g. [45]. In particular, some double-exited levels of He and Li atoms are resonant to the powerful Se^{+24} 20.9 nm X-ray laser. Two transition schemes have been considered by us: (i) He: $1\,^1S \rightarrow 2s2p\,^1P \rightarrow 2\,^1S$ (the Stokes wavelength 32.2 nm), and (ii) Li: $1s^2 2s \rightarrow [1s(2s2p)^1P]^2P \rightarrow 1s^2 3s^2\,S$ (the Stokes wavelength 22.3 nm). Besides generating two more coherent X-ray lines, these processes reveal some new physics.

Table 1. Frequency transformations of XRL radiation by four-wave mixing with optical lasers in plasma. E_{ion} is the ionization potential of the ion, λ is the output wavelength, K is the coefficient for Eq. (6).

Medium E_{ion}	Lasers	K (cm^4/W^4)	λ
1. *Ar VIII 143 eV*	Ge^{22+} 232.24 Å – *Nd 1.06 μm*	1.7×10^{-28}	117.41 Å
2. *K IX 176 eV*	Se^{24+} 209.78 Å – *Nd 0.53 μm*	1.5×10^{-28}	106.99 Å
3. *Ca X 271 eV*	Se^{24+} 206.38 Å – *Nd 0.53 μm*	7.7×10^{-27}	105.24 Å
4. *K IX 176 eV*	Se^{24+} 206.38 Å – *Nd 0.266 μm*	3.0×10^{-27}	107.35 Å
5. *K IX 176 eV*	C^{5+} 182.173 Å – *Nd 0.263 μm*	7.6×10^{-32}	94.35 Å
6. *Ca X 271 eV*	Y^{29+} 154.95 Å – *Nd 0.532 μm*	1.1×10^{-30}	78.62 Å
7. *V XIII 336 eV*	Ag^{37+} 99.36 Å – *Nd 1.06 μm*	3.7×10^{-30}	49.91 Å
8. *Cu XIX 670 eV*	Ta^{45+} 44.83 Å – *Nd 1.06 μm*	6.7×10^{-33}	22.46 Å
9. *Ca XV 894 eV*	C^{5+} 182.173 Å – *KrF 0.2484 μm*	2.9×10^{-29}	94.55 Å
10. *Ar XIII 686 eV*	Se^{24+} 209.78 Å – *KrF 0.2484 μm*	1.7×10^{-32}	109.51 Å
11. *Ar VIII 143 eV*	Ge^{22+} 236.26 Å + 232.24 – *Nd 1.053 μm*	8.0×10^{-29}	118.43 Å
	Ge^{22+} 236.26 Å + 232.24 – *Nd 0.2633 μm*	1.8×10^{-28}	122.57 Å
	Ge^{22+} 236.26 Å + 232.24 – *Nd/St 1.872 μm*	5.8×10^{-25}	117.85 Å
12. *MnXV 435 eV*	Ta^{45+} 44.83 Å + Se^{24+} 206.38 – *Nd 0.5265 μm*	1.8×10^{-28}	37.09 Å

Our estimates of small-signal gain have shown that significant Stokes output requires pump intensity of the order of $10^{12} - 10^{14}\ W/cm^2$, which is feasible. At the same time, such intense X-rays can fully ionize a medium within a fraction of the XRL pulse duration. Thus, effective Stokes generation can take place only at the leading edge of the laser pulse, before the full ionization sets in.

Our numerical solutions of Maxwell-Bloch equations for X-ray SERS [33,34] in Li show two distinct stages of this Raman process, the exponential growth and the saturation. In the exponential region, the Stokes pulse width is constant and its peak coincides with the leading edge of the pump pulse whose velocity is limited by the photoionization to be smaller than the speed of light in vacuum. In the saturation region, the Stokes pulse intensity is almost constant, while the pulse width increases linearly with the distance z. The approximate analytical model developed by us yielded, in particular, an estimate of the optimal length of the Raman medium; e. g., for the attainable XRL pulse energy of 300 μJ, the optimal focusing for $N = 10^{18}\ cm^{-3}$ is $b = L \approx 5.65\ cm$, and the total exponential gain at the cell end is $GL \approx 23$, L being the length of the medium.

One of our major results is the finding that X-ray SERS can significantly inhibit the photoionization of the media and lead to the formation of soliton-like pulses and precursors at the Stokes frequencies. Numerical solution for the intensities and populations for SERS in He shows that the coherent SERS significantly inhibits the photoionization of neutral

atoms by X-ray radiation. This inhibition is due to the fact that a significant portion of neutral atoms ends up being "parked" at the upper excited level, whose photoionization cross section is very small. As a result, "ionization-front stimulated Raman" (IFSR) soliton is formed. While the trailing edge of the Stokes pulse travels with the same velocity as the photoionization front, its leading edge travels much faster, with the velocity of light in the neutral media. The length of such an almost rectangular pulse increases linearly with the distance traveled in media [35]. Therefore, in the X-ray IFSR soliton we have a strong "Stokes precursor", which arrives at the end of the cell significantly ahead of the pumping pulse and can be used for measurements and for "pilot warning" of the trailing photoionization front. For propagating distance of 10 cm in *Li*, pump intensity of 10^{12} *W/cm*^{2} and $N=10^{18}$ *cm*$^{-3}$ (~ *0.1 atm* at T=800K) the "warning time" is ~ *100 ps*.

5. Conclusion

Born more than 25 years ago, X-ray nonlinear optics has experienced a fast theoretical development for the last six years as resonant XRNO. The suitable media have been identified and estimated for a large variety of resonant XRNO effects with virtually all available XRL lines of substantial intensity. Realized experimentally, these effects will result, in particular, in a drastic increase in the number of coherent X-ray sources, including shorter-wavelength and tunable sources. The quantitative estimates show that the majority of the effects considered are feasible with contemporary X-ray laser and plasma technology. Now, the field is ready for experiments.

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