# Generation of attosecond X-ray laser pulses

Riccardo Tommasini and Ernst Fill

***Abstract*— We propose to generate as-X-ray laser pulses by beating of two or more X-ray laser lines with a frequency separation in the range of $10^{15}$ Hz. We focus on nickel-like X-ray lasers, some of which have a few almost equidistant laser gain lines with an appropriate difference frequency. It is shown that in the case of three or more lines, these can be phase-locked by means of a Langmuir wave generated in the gain medium at a suitable electron density.**

***Index Terms*— Attosecond pulses, laser-plasmas, X-ray laser, plasma wave.**

## I. Introduction

In recent years the generation of as pulses in the soft X-ray region has advanced to a stage at which applications of these pulses in many fields of research, such as chemistry, molecular and atomic physics, solid state physics become possible [1]–[3]. First experiments achieving attosecond control of electronic processes and demonstrating time-resolved atomic inner-shell spectroscopy have already been carried out [4], [5]. In such experiments as-soft X-ray pulses are generated by focusing few-cycle laser pulses in a gaseous medium and filtering out a range of the generated harmonics. The synchronization of the generated soft X-ray pulse with the driving laser makes possible pump-probe experiments on a few-fs or even as time scale.

A limitation of these generation techniques is the very small number of photons contained in the X-ray pulse. Even if the harmonics applied are in the plateau region of the harmonic generation spectrum, the emission is only an extremely small fraction of the pump laser emission and, thus experiments require a large number of shots to be accumulated.

In view of this problem it would be desirable to generate as-X-ray pulses with an X-ray laser, in which case a significantly higher number of photons would be available. Today many saturated soft X-ray lasers at wavelengths ranging down to about 6 nm are available using electron-collisional excitation in neon- or nickel-like ions [6]. These lasers typically emit energies in the range of tens of micro–joules.

Unfortunately, however, it seems very difficult to scale soft X-ray lasers down to very short pulse durations. Increasing the laser line width reduces the gain and thus prevents saturation. On the other hand, below saturation, the line width scales as the inverse square root of the gain length product (see, e.g. Ref. [7]), and this spectral narrowing is acting towards elongation of the pulse duration. In the case of inhomogeneous broadening one would expect rebroadening above the onset of saturation. This effect however has never been observed so far and would, in any case, only restore the inhomogeneous component of the line width to its original value, with no significant shortening of the transform limited pulse duration. The shortest pulse duration reported to date is 2 ps, emitted from a nickel-like Ag laser at 13.9 nm [8], and is not far from the transform limited value for the pulse durations, given the typical line widths of this x-ray lasers.

In this paper we point out that the emission of attosecond soft-X-ray laser pulses may be possible by taking advantage of the spectral features of collisionally excited X-ray lasers: the neon- or nickel-like levels between which inversion is achieved show a fine structure resulting from the different projections of the electron angular momentum onto the one of the core. Moreover a number of X-ray lasers emit not only on the primary electron collisionally pumped transition but also on a transition pumped partially by photopumping. For example in neon-like lasers the main transition is the 3p $^1S_0 \rightarrow$ 3s $^1P_1$ (in LS-notation), but several other transitions, e.g. J = 1–1 and J = 2–1 transitions also show gain. In addition gain is also predicted for the photopumped 3d–3p line [9]. Similarly in nickel-like lasers the dominating transition is the 4d$^1S_0 \rightarrow$ 4p$^1P_1$ transition but a number of other singlet and triplet lines and the photopumped 4f $\rightarrow$ 4d lines exhibit gain [10]. Notice that, in the case of Mo, both the 18.9 nm 4d $\rightarrow$ 4p transition and the photo pumped 22.6 nm 4f $\rightarrow$ 4d line have been experimentally observed to show inversion at the same time [11].

Taking advantage of these facts one realizes that beating of two closely spaced transitions leads to a series of attosecond pulses, with a width of the inverse beat frequency and spaced by the same time. However, the situation is more favorable if three or more lines have an almost equal frequency spacing. In this case a train of pulses with a gap between each other is generated, provided the phases are appropriately locked. In this paper we concentrate specifically on the case of nickel-like molybdenum which has its main gain line at 18.9 nm, but which exhibits a number of other lines with high gain.

## II. Generating As Pulse trains

Beating two lines with wavelengths $\lambda_1$ and $\lambda_2$ results in a series of pulses with a full half width of

$$\tau \approx \lambda_1\lambda_2/[2c(\lambda_2-\lambda_1)] \tag{1}$$

While such a pulse train may already be useful in certain experiments, the situation is more favorable if a series of equally spaced lines is considered. For $n$ lines with wavelengths $\lambda_1$, $\lambda_2$,...,$\lambda_n$, spaced by a wavelength separation $\Delta\lambda$ the resulting pulse duration is given by

$$\tau \approx \lambda^2/(nc\Delta\lambda) \tag{2}$$

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where $\lambda$ is an average wavelength. The peak intensity of the pulses is increased by a factor of $n^2$.

Equation(2) is valid only if the amplitudes of the fields are equal and the phases are ideally locked. However, it can be shown, that the regular structure of the pulses does not depend very much on the amplitudes at the different wavelengths being equal, but is very sensitive to the phase relation of the modes [12]. In the case of harmonics, propagation effects can lead to phase locking and thus to a train of isolated as pulses [1]. Moreover, the "single cycle" regime of harmonic generation leads to increased coherence and self-phase-matching [13], [14]. For X-ray lasers such effects can not be invoked and phase-locking must be achieved by other means. In the following we propose to achieve phase locking of three lines by coupling the laser lines to a strong plasma wave. The analysis will show that conditions can be found under which the plasma wave optimally locks the phases of the individual lines and thus a regular pulse train is achieved.

The wavelengths and gain coefficients for gain lines in nickel-like molybdenum have been investigated by Nilsen [10]. His calculations show 7 gain lines near 20 nm with gain coefficients above 100 $cm^{-1}$. The electron densities at maximum gain are between $10^{20}$ and $10^{21}$ $cm^{-3}$.

Inspection of the calculated spectrum reveals that the dominating lines at 18.9 and 22.0 nm which are predicted to have similar gain. Moreover, there is a "triplet" of gain lines with wavelengths of 24.1, 24.5 and 24.8 nm respectively, with the latter one predicted to have the highest gain.

Notice that the self photo-pumped line predicted at 22 nm, has been measured later on to be actually centered at 22.6 nm [15]. Spectra showing inversion on both lines have been measured even when using high-repetition, i.e. 10 Hz, rate pumping [11]. Simulations shown in Ref. [11] calculating the gain versus time and space, indicate perfect temporal overlap of the gain for the two lines, at least on a time scale much smaller than the ps time scale characteristic of the emission on these two lines. Thus indicating a very good temporal overlap of the two beating lines.

Using eq.1 it is determined that beating of the 18.9 and 22.6 nm lines results in a series of pulses with a duration of 192 as. Obviously in this case the phases of the lines are irrelevant, and the particular choice of phases just shifts the time axis of the pulses. Fig.[1] shows the intensity obtained in the case of beating of these lines.

Two-line x-ray lasing (4d–4p transitions) is in principle possible in all the nickel-like ions. To restrict ourself on the experimentally demonstrated lasing lines we will mention a few more examples. Particularly interesting is the case of nickel-like Dy. The two 4d–4p lasing lines for nickel-like Dy have shown to simultaneously operate in saturation at 5,86 and 6.37 nm [16]. This means that the two lasing lines can operate at high stability, producing trains of pulses with duration of 122 as. The cases of nickel-like Tb [17] and Yb [18] are also worth to mention, showing two-line x-ray lasing at 5.9 and 6.7 nm and at 5.026 and 5.609 nm respectively. The beating of this two lines will result in a pulse duration of 82 and 80 as, respectively.

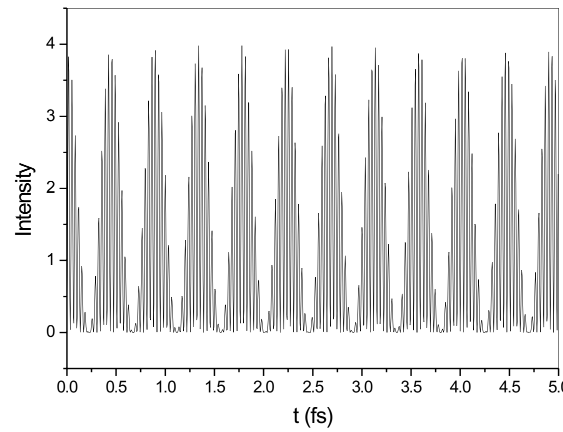


Fig. 1. Intensity pattern of Mo X-ray laser emission consisting of the two gain lines at 18.9 and 22.6 nm

## III. Locking the phases of three X-ray laser lines

The situation in which three or more lines have an almost equal frequency spacing is extremely interesting. In this case a train of pulses with a gap between each other is generated, provided the phases are appropriately locked. Concerning this possibility, we will concentrate specifically on the case of nickel-like molybdenum which will be the prototype for our proposed mechanisms of phase locking.

For the gain line "triplet", straightforward application of eq.(2) predicts a pulse duration of 2.2 fs while the individual pulses are separated by 6.6 fs. However, if the modes are not ideally phased, an irregular intensity pattern with a larger pulse duration, a lower peak intensity will be generated. Of course, the pattern will still be periodic with a period of 6.6 fs.

We propose to lock the phases of X-ray laser lines by driving a strong Langmuir wave at the beat frequency of the X-ray laser lines in a plasma of appropriate density. Most efficiently this can be achieved in the X-ray laser medium itself, if the beat frequency is equal to the plasma frequency. For this to be possible the beat frequency must occur at a density at which the X-ray laser lines exhibit gain. We will show below that under appropriate conditions the plasma wave couples the laser waves resulting in optimum phase locking for the generation of ultrashort pulses. Driving a resonant plasma wave by two laser frequencies has been proposed for heating a plasma. Furthermore it is used in the beat wave accelerator to accelerate electrons to high energies [19]–[21].

We consider two lasers with frequencies $\omega_l$ and $\omega_{l-1}$ irradiating an underdense plasma. If the plasma frequency $\omega_p$ obeys the condition

$$\omega_l - \omega_{l-1} = \omega_p + \Delta_l \quad (3)$$

where $\Delta_l$ is a small frequency mismatch between the beat wave and the plasma frequency, the plasma wave is strongly excited and driven to a high amplitude. Interaction of the lasers with the plasma wave generates sidebands spaced by the plasma frequency. The third laser line is generated from noise by parametric interaction in the plasma. In the following analysis the evolution of the phases of the waves participating in the interaction are investigated.

The coupled equations for the amplitude $\varphi$ of the electrostatic potential of the Langmuir wave and the vector potential amplitudes $A_l$ of the laser fields are obtained from the Maxwell- and the material equations under the slowly varying envelope approximation. They are given by [20]

$$\left(\frac{\partial}{\partial t}+\gamma\right)\varphi=\kappa\sum_l A_l A_{l-1}^{*}\mathrm{e}^{-\mathrm{i}\Delta\mathrm{t}} \tag{4a}$$

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial z}\right)A_l=(\kappa/l)\left[A_{l+1}\varphi^{*}\mathrm{e}^{-\mathrm{i}\Delta\mathrm{t}}-\mathrm{A}_{l-1}\varphi\mathrm{e}^{\mathrm{i}\Delta\mathrm{t}}\right] \tag{4b}$$

In these equations $\gamma$ is the damping coefficient of the Langmuir wave, $\kappa=e\omega_p^2/2mc^2$ and $l=\omega_l/\omega_p$. The physical situation has been simplified by assuming only a single detuning parameter $\Delta$. Plasma dispersion and relativistic effects are neglected. These equations have been used to investigate the distribution of laser energy into Raman sidebands generated by the plasma wave [20]–[22].

The system of equations 4 is simplified by restricting the analysis to only three laser waves with amplitudes $A_{l-1}, A_l$ and $A_{l+1}$, justified by the fact that only these three waves have gain. The boundary value problem, in which the laser intensities are only space, but not time-dependent is considered. This situation applies to an X-ray laser if the lasing pulse is much longer than the separation of the ultrashort pulses. We introduce phenomenological intensity gain coefficient $g$, assumed equal for the three laser lines.

With these simplifications and alterations eqs.(4) become

$$\left(\frac{\partial}{\partial t}+\gamma\right)\varphi=\sum_l^{l+1}A_l A_{l-1}^{*}\,\mathrm{e}^{-\mathrm{i}\Delta\mathrm{t}} \tag{5a}$$

$$\frac{\partial}{\partial z}A_l=(\kappa/l\,c)\,\left[A_{l+1}\varphi^{*}\mathrm{e}^{-\mathrm{i}\Delta\mathrm{t}}-\mathrm{A}_{l-1}\varphi\mathrm{e}^{\mathrm{i}\Delta\mathrm{t}}\right]+(g/2)A_l \tag{5b}$$

$$\frac{\partial}{\partial z}A_{l+1}=-(\kappa/l\,c)\,A_l\varphi\mathrm{e}^{\mathrm{i}\Delta\mathrm{t}}+(\mathrm{g}/2)\mathrm{A}_{l+1} \tag{5c}$$

$$\frac{\partial}{\partial z}A_{l-1}=(\kappa/l\,c)\,A_l\varphi^{*}\mathrm{e}^{-\mathrm{i}\Delta\mathrm{t}}+(\mathrm{g}/2)\mathrm{A}_{l-1} \tag{5d}$$

In eqs. 5c and 5d the numbers $l$+1 and $l$-1 are approximated by $l$. Note that the sum in 5a now runs only from $l$ to $l$+1, thus considering only the three lines assumed to have gain.

Since the $A_l$ are not time-dependent eq.(5a) can be solved independently yielding

$$\varphi=\frac{\kappa}{\gamma-i\Delta}Be^{-i\Delta t}, \tag{6}$$

where the sum in eq.(5a) has been abbreviated by $B=A_lA_{l-1}^{*}+A_{l+1}A_l^{*}$. Equation (6) shows that the plasma wave grows in space as the laser intensity and is driven at the beat frequency of the lasers, not at the plasma frequency. The evolution of the laser amplitudes and phases can now be elaborated by inserting $\varphi$ in eqs. (5b,5c,5d). Setting $A_l=a_l\mathrm{e}^{\mathrm{i}\theta_l}$; $A_{l+1}=a_{l+1}\mathrm{e}^{\mathrm{i}\theta_{l+1}}$; $A_{l-1}=a_{l-1}\mathrm{e}^{\mathrm{i}\theta_{l-1}}$, inserting in eqs. (5b, 5c, 5d) and equalizing the real and imaginary parts of the equations, separately, yields for the amplitudes

$$\frac{\partial}{\partial z}a_l=K\left[\gamma\left(a_{l+1}^2a_l-a_la_{l-1}^2\right)-Re\left(2i\Delta\;a_{l+1}a_la_{l-1}\mathrm{e}^{\mathrm{i}\delta\theta}\right)\right]+(g/2)a_l \tag{7a}$$

$$\frac{\partial}{\partial z}a_{l+1}=-K\left\{\gamma a_l^2a_{l+1}+Re\left[(\gamma+i\Delta)a_l^2a_{l-1}\mathrm{e}^{-\mathrm{i}\delta\theta}\right]\right\}+(g/2)a_{l+1} \tag{7b}$$

$$\frac{\partial}{\partial z}a_{l-1}=K\left\{\gamma a_l^2a_{l-1}+Re\left[(\gamma-i\Delta)a_l^2a_{l+1}\mathrm{e}^{-\mathrm{i}\delta\theta}\right]\right\}+(g/2)a_{l-1} \tag{7c}$$

and for the phases

$$\frac{\partial}{\partial z}\,\theta_l=-K\left[\Delta(a_{l+1}^2+a_{l-1}^2)+Im(2i\Delta a_{l+1}a_{l-1}\mathrm{e}^{\mathrm{i}\delta\theta})\right] \tag{8a}$$

$$\frac{\partial}{\partial z}\,\theta_{l+1}=-K\left\{\Delta a_l^2+Im\left[(\gamma+i\Delta)\frac{a_l^2a_{l-1}}{a_{l+1}}\mathrm{e}^{-\mathrm{i}\delta\theta}\right]\right\} \tag{8b}$$

$$\frac{\partial}{\partial z}\,\theta_{l-1}=K\left\{-\Delta a_l^2+Im\left[(\gamma-i\Delta)\frac{a_l^2a_{l+1}}{a_{l-1}}\mathrm{e}^{-\mathrm{i}\delta\theta}\right]\right\} \tag{8c}$$

In eqs.(7) and (8) $K=\kappa^2/[l\,c\,(\gamma^2+\Delta^2)]$ and $\delta\theta=\theta_{l+1}-2\theta_l+\theta_{l-1}$.

The investigation of eqs.(7) and (8) becomes particularly simple if two limiting cases are considered:

1. Linear damping of the plasma wave is dominating, i.e.$\gamma\gg\Delta$. This case leads to ideal phase locking: Neglecting $\Delta$ in eqs. (8) yields

$$\frac{\partial}{\partial z}\theta_l=0 \tag{9a}$$

$$\frac{\partial}{\partial z}\theta_{l+1}=-KIm\left(\gamma\frac{a_l^2a_{l-1}}{a_{l+1}}e^{-i\delta\theta}\right) \tag{9b}$$

$$\frac{\partial}{\partial z}\theta_{l-1}=KIm\left(\gamma\frac{a_l^2a_{l+1}}{a_{l-1}}e^{-i\delta\theta}\right) \tag{9c}$$

and for $\delta\theta=0$ the derivatives of all phase angles are zero! However, this would also be the case for $\delta\theta=\pi$. In order that the system locks to the condition $\delta\theta=0$ one needs $a_{l+1}>a_{l-1}$. Indeed in this case the equation for $\delta\theta$ reads

$$\frac{\partial}{\partial z}\delta\theta=-K\gamma\frac{a_l^2}{a_{l+1}a_{l-1}}\left(a_{l+1}^2-a_{l-1}^2\right)\;\sin\delta\theta \tag{10}$$

having decaying solution

$$\delta\theta(z)\propto\mathrm{Arccot}\left[\exp\left(\mathrm{K}\gamma\frac{\mathrm{a}_l^2}{\mathrm{a}_{l+1}\mathrm{a}_{l-1}}\left(\mathrm{a}_{l+1}^2-\mathrm{a}_{l-1}^2\right)\mathrm{z}\right)\right] \tag{11}$$

which means $\delta\theta$ goes to zero for any initial condition of the phases. Note that $\delta\theta$=0 implies $\theta_{l+1}-\theta_l=\theta_l-\theta_{l-1}$ i.e. the phases are equidistant. The particular choice of the phases is not relevant since it only results in a shift along the time axis. The time evolution of the intensity for $\delta\theta$=0 is shown in fig.(2a) for the case of equal normalized intensities. It is seen that the peak intensity reaches a value of 9 times the intensity of the individual modes. In between the pulses a small maximum of about 10% of the main maximum is observed, resulting from the fact that only three modes are locked.

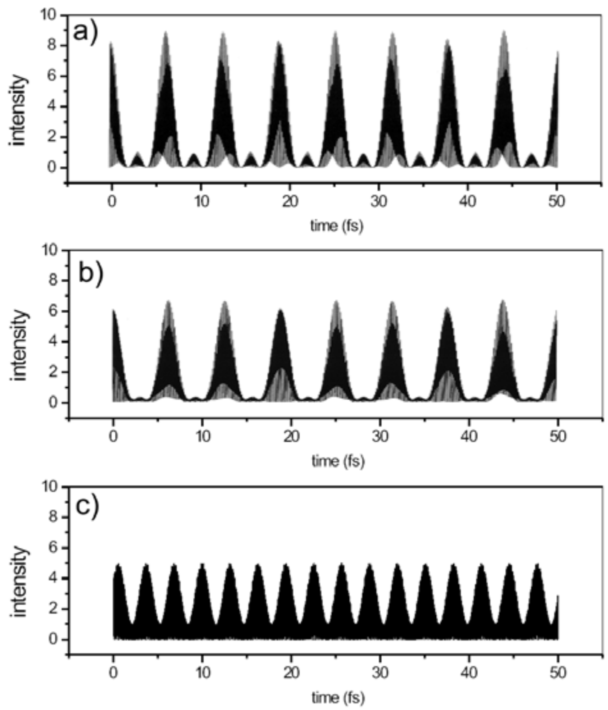


Fig. 2. Pulse train obtained by beating of three molybdenum X-ray laser lines. Three cases of phase locking are displayed. a) $\gamma \gg \Delta$ and $\delta\theta = 0$; b) $\gamma \gg \Delta$ and $\delta\theta = \pi$; c) $\Delta \gg \gamma$ and $\delta\theta = \pi$. The first two cases represent ideal phase locking for the three modes. The last case exhibits about the same pulse duration at twice the repetition rate but with lower intensity.

Analogously the time evolution of the intensity for $\delta\theta$=$\pi$ is shown in fig.(2b) for equal normalized intensities. In this case the peak intensity reaches a value of about 6.75 times the intensity of the individual modes, while the secondary maxima are lower in intensity, reaching only about 2% of the main maxima.

2. The second case implies detuning of the beat frequency dominating over damping of the Langmuir wave, i.e. $\Delta \gg \gamma$. Again phase locking is obtained, but the phases lock to the condition $\delta\theta = \pi$. The temporal evolution of the intensity for this case is illustrated in fig.(2c). It is seen that the pulses have about the same duration as in the previous case, but they occur at twice the rate and the peak intensity is only a factor of about two higher than the average intensity. Thus, this case is less favorable for achieving isolated ultrashort pulses.

## IV. Application to X-ray lasers

The foregoing analysis has shown that, besides trains of attosecond pulses obtained from the beating of two lasing lines, under special conditions it may be possible to force an X-ray laser to emit a series of spaced ultrashort pulses from the beating of three or more lasing lines, the pulse duration being in the attosecond range. In the case of the nickel-like Mo laser this requires lasing on three gain lines approximately equidistant and a plasma density matching the plasma frequency to the beat frequency of the lasers. For the Mo gain lines investigated this density is $2.8{\times}10^{20}\mathrm{cm}^{-3}$, which is quite favorable to generate high gain. In fact the simulations of Ref. [10] show that for the main gain lines the gain peaks at $8.9{\times}10^{20}\mathrm{cm}^{-3}$. It is to be expected that the gain lines in question have maximum gain at a somewhat lower electron density.

Notice that, while in the specific case under study of a Mo x-ray laser, the pulse duration is about 2 fs, the expected pulse duration will drop to the as range by scaling down the central wavelength using a higher Z nickel- or neon-like ion as the active medium.

The question arises about how closely has the electron density to be matched to the critical density of the beat wave in order to fulfill the condition for optimum phase locking: $\gamma \gg \Delta$. We take only collisional damping into account since Landau damping is absent due to the high velocity of the plasma wave. The collisional damping rate is close to the collision frequency between ions and electrons. Approximately one obtains [22]

$$\frac{\gamma}{\omega_p} = \frac{\ln \Lambda}{2\pi N_e \lambda_D^3}, \qquad (12)$$

where $\ln\,\Lambda$ is the Coulomb logarithm, $N_e$ is the electron density and $\lambda_D$ is the Debye length of the plasma. For the X-ray laser plasma parameters k$T_e$ = 300 eV, $N_e$ = $2.8{\times}10^{20}\mathrm{cm}^{-3}$, ln $\Lambda$=10, one obtains $\gamma/\omega_p \approx 2.6 \times 10^{-2}$. Thus, to fulfill the condition for optimum mode coupling, the electron density has to be controlled with an accuracy of about one percent. While this accuracy may appear discouraging, we notice that this represents the ideal condition only. Indeed since phase locking will happen also in the case of detuning of the beat frequency dominating over damping of the Langmuir wave (see case 2 of the previous section), the condition of matching the electron density to the critical density of the beat wave is greatly relaxed and not crucial.

In our analysis we assumed the different lasing lines having the same polarization. This is justified on the basis of experimental evidences that a good degree of intrinsic linear polarization can develop in the amplified spontaneous emission build up of an x-ray laser [23]–[25], even though the mechanism for this is not completely understood [26]. More recently Gavrilenko and Oks [27] have shown that the polarization of the x-ray lasing line can be controlled via dressing by an elliptically polarized electric field of an optical laser. This would open the possibility for the polarization of the different beating lasing lines to be actively controlled.

## V. Conclusions

We have shown that the spectral emission characteristics of electron collisionally pumped soft X-ray lasers allows generation of pulse trains with attosecond or few-fs time durations. This is achieved either by beating of two X-ray laser lines at different wavelengths or by superposition of three (or maybe more) equidistant X-ray laser lines. In case of three lines phase locking of the different frequencies can be achieved by generating a plasma wave at the beat frequency of the laser lines. Different cases of plasma wave damping and detuning between beat frequency and plasma frequency lead to phase locking with different intensity patterns.

The possibility of generating ultrashort X-ray laser pulses in the as and few-fs range is very exciting in view of possible experiments. It opens up the possibility to perform pump-probe experiments with X-ray lasers on an attosecond or few-femtoseconds time scale.

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