Towards megajoule x-ray lasers via relativistic four-photon cascade in plasma

V. M. Malkin and N. J. Fisch
Department of Astrophysical Sciences, Princeton University, Princeton, New Jersey USA 08540

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A theoretically highly efficient mechanism, operating at high laser intensities and powers, is identified for spectral transferring huge laser energies to shorter ultraviolet and x-ray wavelengths. With megajoule laser energies currently available at near-optical wavelengths, this transfer would, in theory, enable megajoule x-ray lasers, a huge advance over the millijoules x-ray pulses produced now. In fact, enabling even kilojoule x-ray lasers would still be a fantastic advance, and a more likely achievable one, considering practical experimental inefficiencies.

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I. INTRODUCTION

The highest energy intense laser pulses are currently produced in the near-optical range through chirped pulse amplification mediated by material gratings [1]. However, material gratings cannot be employed at much shorter wavelengths, so that available energies of intense laser pulses dramatically drop at ultraviolet and x-ray wavelengths. In particular, the most energetic short x-ray pulses, currently produced by giant free-electron lasers, are now only in the millijoule range [2]. Hence, an efficient transfer of laser energies from near-optical wavelengths, where megajoule laser energies are available [3], to deep ultraviolet and x-ray wavelengths would open up new research and technological frontiers.

The desired spectral energy transfer cannot be accomplished through methods of high harmonic generation in gases [4,5] or crystals [6], because gases and crystals cannot tolerate ultrahigh intensities. Huge intensities can be tolerated in plasma. Several techniques do exploit the properties of plasma to generate very high intensity pulses through resonant three-wave interactions, like Raman backscattering [7–12], Brillouin scattering [13–17], or magnetized low-frequency scattering [18]. However, these techniques cannot be adapted to produce the significant frequency upshifts contemplated here.

Frequency upshifts of laser energy could be achievable via resonant four-photon interactions in plasma, where two photons are scattered into a higher frequency photon and a lower frequency photon [19]. However, there are severe impediments to this approach at high laser powers. Here we show how to overcome these impediments. Then cascading a few stages, each of which nearly doubles the photon frequency, might transfer huge laser energies from optical to ultraviolet wavelengths; cascading 10 stages would, in theory, dramatically and efficiently convert optical wavelength laser energy to nanometer wavelengths.

The classical synchronism conditions for the resonant four-photon scattering in plasma are as follows:

$$\boldsymbol{k}_1+\boldsymbol{k}_2=\boldsymbol{k}_3+\boldsymbol{k}_4, \quad \omega_1+\omega_2=\omega_3+\omega_4,$$
$$\omega_j=\sqrt{k_j^2c^2+\omega_e^2}, \quad \omega_e=\sqrt{4\pi n_0 e^2/m}, \tag{1}$$

where c is the speed of light in vacuum, m is the electron rest mass, $-e$ is the electron charge, n_0 is the electron density, ω_e is the electron plasma frequency, ω_j are laser frequencies, and $\boldsymbol{k}_j$ are laser wave vectors in plasma.

Intense laser pulses could be coupled in plasma through the relativistic electron nonlinearity. So far, the most theoretical attention has been devoted to the very degenerate case of collinear laser pulses. For the collinear pulses, the synchronism conditions Eq. (1) have only trivial solution $\boldsymbol{k}_1=\boldsymbol{k}_3$, $\boldsymbol{k}_2=\boldsymbol{k}_4$ or $\boldsymbol{k}_1=\boldsymbol{k}_4$, $\boldsymbol{k}_2=\boldsymbol{k}_3$. Thus, there are not four but only two laser pulses, and such a four-photon interaction does not produce a real four-photon scattering but only nonlinear frequency shifts.

For moderately noncollinear pulses in a very undercritical plasma, $\omega_e \ll \omega_j$, $k_{j\perp} \gg \omega_e/c$, $\boldsymbol{k}_{j\perp}\cdot\mathbf{K}=0$, $\mathbf{K}\equiv\boldsymbol{k}_1+\boldsymbol{k}_2=\boldsymbol{k}_3+\boldsymbol{k}_4$, the frequency resonance condition reduces to $k_1+k_2\approx k_3+k_4$, like in vacuum, where the $\boldsymbol{k}_j$ vectors trace an ellipsoidal manifold, Fig. 1. Thus, many photon pairs having the same $\mathbf{K}$ and ω, $2\omega\equiv\omega_1+\omega_2=\omega_3+\omega_4$ can experience real four-photon scattering.

We show here how the relativistic four-photon scattering of moderately noncollinear laser pulses opens up unprecedented possibilities of highly efficient spectral transfers of huge laser energies to shorter wavelengths. Moreover, such transfers can be accomplished at very high powers P, exceeding the critical power $P_{\rm cr}$ of relativistic self-focusing of laser pulses in plasma [20–23],

$$P_{\rm cr}=m^2c^5\omega^2/e^2\omega_e^2\approx 17\,(\omega/\omega_e)^2\,\mathrm{GW}. \tag{2}$$

To show this, we first need to get a sufficiently general three-dimensional description of the relativistic four-photon coupling in plasma, which has not yet been available in a form suitable for addressing this problem.

II. RELATIVISTIC FOUR-PHOTON COUPLING

We begin with the Maxwell equations in Coulomb gauge and Hamilton-Jacobi equation for electron motion in electromagnetic fields [24] (kinetic effects are neglected, because all

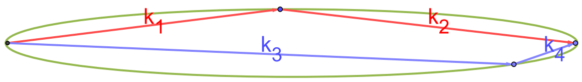

FIG. 1. Resonant wave-vector ellipsoid.

the beating phase velocities and electron quiver velocities considered here are much larger than electron thermal velocities):

$$\boldsymbol{H} = \nabla \times \boldsymbol{A}, \quad \boldsymbol{E} = -\partial_{ct}\boldsymbol{A} - \nabla\Phi, \quad \nabla\cdot\boldsymbol{A} = 0, \tag{3}$$

$$\Box \boldsymbol{A} \equiv \partial_{ct}^2 \boldsymbol{A} - \Delta \boldsymbol{A} = 4\pi \boldsymbol{J}/c - \partial_{ct}\nabla\Phi, \tag{4}$$

$$\boldsymbol{J} = -en\boldsymbol{P}/\sqrt{m^2 + P^2/c^2}, \quad \Delta\Phi = 4\pi e(n - n_0), \tag{5}$$

$$\boldsymbol{P} = e\boldsymbol{A}/c + \nabla S, \quad \partial_t S = e\Phi - c\sqrt{m^2c^2 + P^2} + mc^2. \tag{6}$$

By introducing dimensionless electromagnetic potentials and electron momentum, and rescaling the action function S by the factor mc^2,

$$\boldsymbol{A} = mc^2\boldsymbol{a}/e, \quad \Phi = mc^2\phi/e, \quad \boldsymbol{P} = mc\boldsymbol{p}, \quad S = mc^2 s, \tag{7}$$

the equations can be presented in the forms

$$c^2\Box\boldsymbol{a} = -\boldsymbol{p}(1 + p^2)^{-1/2}\big(\omega_e^2 + c^2\Delta\phi\big) - c\partial_t\nabla\phi, \tag{8}$$

$$\nabla\cdot\boldsymbol{a} = 0, \quad \boldsymbol{p} = \boldsymbol{a} + c\nabla s, \quad \partial_t s = \phi - \sqrt{1 + p^2} + 1. \tag{9}$$

For mildly relativistic electron quiver velocities in laser pulses, $a \ll 1$, Eqs. (8) and (9) can be expanded in powers of the small parameter a. To calculate the four-photon coupling, the expansion should include terms up to cubic in a. With laser beatings well off the plasma wave resonance, the leading term of the electrostatic potential ϕ expansion is quadratic in a, so that the cubic in a expansion of Eq. (8) is

$$\big(c^2\Box + \omega_e^2\big)\boldsymbol{a} = \boldsymbol{a}\big(a^2\omega_e^2/2 - c^2\Delta\phi\big) - c\big(\omega_e^2\nabla s + \nabla\partial_t\phi\big). \tag{10}$$

Equation $\nabla\cdot\boldsymbol{a} = 0$ can be used to exclude s from Eq. (10). In a uniform plasma, $\nabla\omega_e = 0$, it gives

$$\omega_e^2 s + \partial_t\phi = \Delta^{-1}\nabla\cdot\big[\boldsymbol{a}\big(a^2\omega_e^2/2 - c^2\Delta\phi\big)\big]/c, \tag{11}$$

$$\big(c^2\Box + \omega_e^2\big)\boldsymbol{a} = (1 - \nabla\Delta^{-1}\nabla\cdot)\big[\boldsymbol{a}\big(a^2\omega_e^2/2 - c^2\Delta\phi\big)\big]/c. \tag{12}$$

Equations (11) and $\partial_t s = \phi - \sqrt{1 + p^2} + 1$ can be used now to exclude ϕ from Eq. (12),

$$\big(\partial_t^2 + \omega_e^2\big)\phi = a^2\omega_e^2/2, \tag{13}$$

$$\big(c^2\Box + \omega_e^2\big)\boldsymbol{a} = (1 - \nabla\Delta^{-1}\nabla\cdot)\boldsymbol{a}\big[1 - c^2\Delta\big(\partial_t^2 + \omega_e^2\big)^{-1}\big]a^2\omega_e^2/2. \tag{14}$$

For paraxial laser pulses of frequencies which differences well exceed $\sqrt{\omega_e^2 + k_\perp^2 c^2}$, the beatings of a^2 propagate with speeds close to c. Then the second term in the square brackets in Eq. (14), associated with the electrostatic potential ϕ, nearly exactly compensates the first term in the square brackets, associated with the relativistic variation of electron mass. The compensation significantly reduces the four-photon coupling for paraxial laser pulses. This important effect is missed in calculations of the four-photon scattering probability [25,26] neglecting the relativistic variation of the electron mass.

Apart from their resonant interaction, four laser pulses produce small nonresonant beatings $\delta\boldsymbol{a}$, so that the total field has the form

$$\boldsymbol{a} = \sum_{j=1,\sigma=\pm}^{j=4} \boldsymbol{a}_{\sigma j}\exp[\imath(\boldsymbol{k}_{\sigma j}\boldsymbol{r} - \omega_{\sigma j}t)] + \delta\boldsymbol{a}, \tag{15}$$

$$\boldsymbol{k}_j\cdot\boldsymbol{a}_j = 0, \quad \boldsymbol{a}_{-j} = \boldsymbol{a}_j^*, \quad \boldsymbol{k}_{-j} = -\boldsymbol{k}_j, \quad \omega_{-j} = -\omega_j. \tag{16}$$

Substituting Eq. (15) into Eq. (14) and collecting the resonant terms leads to the following equations for slowly varying canonical amplitudes [27] $b_j = a_j\sqrt{\omega_j}$:

$$\left[\imath\left(\frac{\partial}{\partial t} + \frac{c^2\boldsymbol{k}_j\cdot\nabla}{\omega_j}\right) - \frac{c^2\Delta_{j\perp}}{2\omega_j}\right]b_j = \delta\omega_j b_j + \frac{\partial\mathcal{H}}{\partial b_j^*}, \tag{17}$$

$$\mathcal{H} = Vb_1b_2b_3^*b_4^* + \text{c.c.}, \tag{18}$$

$$\delta\omega_j = \frac{\omega_e^2}{2\omega_j}\left(\sum_{l=1,\sigma=\pm}^{l=4,\sigma l\neq j}\frac{|b_l e_{j,-\sigma l}|^2}{\omega_l}f_{j,-\sigma l} - \sum_{l=1}^{l=4}\frac{|b_l|^2}{\omega_l}\right), \tag{19}$$

$$V = \frac{\omega_e^2(f_{1,2}e_{1,2}e_{-3,-4} + f_{1,-4}e_{1,-4}e_{2,-3} + f_{2,-4}e_{1,-3}e_{2,-4})}{2\sqrt{\omega_1\omega_2\omega_3\omega_4}},$$

$$e_{j,l} = \frac{\boldsymbol{a}_j\cdot\boldsymbol{a}_l}{a_ja_l}, \quad f_{j,l} = \frac{c^2(\boldsymbol{k}_j + \boldsymbol{k}_l)^2}{(\omega_j + \omega_l)^2 - \omega_e^2} - 1 \tag{20}$$

($\Delta_{j\perp}$ is the Laplacian in the plane perpendicular to $\boldsymbol{k}_j$).

III. TRANSVERSE FILAMENTATION INSTABILITY

For a single laser pulse, $b_1 = a\sqrt{\omega}$, $b_2 = b_3 = b_4 = 0$, Eqs. (17)–(20) reduce to the standard cubic nonlinear Schroedinger equation,

$$\left[\imath\left(\frac{\partial}{\partial t} + \frac{c^2k}{\omega}\frac{\partial}{\partial z}\right) - \frac{c^2\Delta_\perp + \omega_e^2|a|^2}{2\omega}\right]a = 0. \tag{21}$$

A spatially uniform solution of this equation,

$$a = a_0\exp(-\imath\Gamma t), \quad \Gamma = |a_0|^2\omega_e^2/2\omega, \tag{22}$$

can experience small transverse modulations

$$\tilde{a} = \exp(-\imath\Gamma t)\{\psi\exp[\imath(\boldsymbol{\kappa r}_\perp - \Omega t)] + \chi^*\exp[-\imath(\boldsymbol{\kappa r}_\perp - \Omega^* t)]\},$$

$$\Omega^2 = c^2\kappa^2/2\omega(c^2\kappa^2/2\omega - 2\Gamma) \equiv \Omega_\kappa^2. \tag{23}$$

At $\kappa < \sqrt{2}\,a_0\omega_e/c$, the modulations are unstable. The largest growth rate, reached at $\kappa = a_0\omega_e/c$, is Γ. Pulses of powers exceeding the critical power Eq. (2) have apertures sufficient to accommodate unstable modulations.

For two laser pulses, such that $|b_2| \approx |b_1|$, $b_3 = b_4 = 0$, $\boldsymbol{e}_{1,-2} \approx 1$, $|\boldsymbol{k}_2 - \boldsymbol{k}_1| \ll k_1$, and $\omega_1 \gg \omega_2 - \omega_1 > \omega_e$, Eqs. (17)–(20) reduce to

$$\left[\imath\left(\frac{\partial}{\partial t} + \frac{c^2\boldsymbol{k}_1\cdot\nabla}{\omega_1}\right) + \frac{\omega_e^2(G|a_2|^2 - |a_1|^2) - c^2\Delta_{1\perp}}{2\omega_1}\right]a_1 = 0,$$

$$\left[\imath\left(\frac{\partial}{\partial t} + \frac{c^2\boldsymbol{k}_2\cdot\nabla}{\omega_2}\right) + \frac{\omega_e^2(G|a_1|^2 - |a_2|^2) - c^2\Delta_{2\perp}}{2\omega_2}\right]a_2 = 0,$$

$$G \approx f_{1,-2} - 1. \tag{24}$$

These equations have a spatially uniform solution

$$a_2 \approx a_1 = a_0 \exp(\imath \Gamma_2 t), \quad \Gamma_2 \approx (G-1)\Gamma \tag{25}$$

with Γ of Eq. (22). Small transverse to $\mathbf{K}$ modulations of this solution satisfy the dispersion equation

$$\left[\left(\frac{\Omega}{\kappa c}-\frac{k_\perp c}{\omega}\right)^2-\frac{\Omega_\kappa^2}{\kappa^2 c^2}\right]\left[\left(\frac{\Omega}{\kappa c}+\frac{k_\perp c}{\omega}\right)^2-\frac{\Omega_\kappa^2}{\kappa^2 c^2}\right] \approx \frac{G^2\Gamma^2}{\omega^2}, \tag{26}$$

where $\boldsymbol{k}_\perp \equiv \boldsymbol{k}_{1\perp} = -\boldsymbol{k}_{2\perp}$ and Ω_κ^2 is given by Eq. (23). As seen from Eq. (26), modulations of pulses 1 and 2 are weakly coupled for $k_\perp^2 \gg G\Gamma\omega/c^2$, a condition satisfied for $(\omega_2-\omega_1-\omega_e)k_\perp^2 c^2/\omega_e^2 \gg |a_0|^2\omega_e$. In the opposite limit, the modulations are strongly coupled and $\Omega^2 \approx \Omega_\kappa^2 \pm G\Gamma\kappa^2 c^2/\omega$. The "+" branch is stabilized for $G>1$, but then the "−" branch gets even more unstable than for a single pulse. Thus, achieving the defocusing nonlinear frequency shift in the uniform solution (25) for $G>1$ does not prevent the transverse filamentation instability. Modulations of collinear laser pulses are necessarily strongly coupled and unstable at total powers exceeding the critical power. (This explains why achieving a defocusing nonlinear frequency shift for collinear laser pulses [28,29] did not help increase to any significant degree the laser power propagating in plasma without filamentation, as was borne out by numerical simulations [29].) In contrast, modulations of noncollinear laser pulses can easily satisfy conditions for the weakly coupled regime and be stable to transverse modulations at each pulse power not exceeding its own critical power. The total power of many stable weakly coupled pulses can then significantly exceed the individual critical powers.

IV. LINEAR GROWTH RATE OF FOUR-PHOTON AMPLIFICATION

Resonant four-photon amplification can be initiated by two pump pulses 1 and 2 and small seed pulse 3. In the paraxial geometry with the axis along $\mathbf{K}$, so that $\boldsymbol{k}_{2\perp} = -\boldsymbol{k}_{1\perp}$, $\boldsymbol{k}_{4\perp} = -\boldsymbol{k}_{3\perp}$, the frequency resonance condition reduces to $k_{1\perp}^2 k_3 k_4 \approx k_{3\perp}^2 k_1 k_2$. For moderately close pump frequencies, $\omega_1 \gg \omega_2-\omega_1 \gg \omega_e$, and moderately large ratio of seed frequencies, $\omega_3 \gg \omega_4$, the pump energy mostly goes into the amplified pulse 3, whose frequency is nearly twice the pump frequencies. When all polarizations are the same, the four-photon coupling coefficient Eq. (20) reduces to

$$V \approx 3\,\omega_e^2 k_{1\perp}^2 \left(2\omega_1\sqrt{\omega_3\omega_4}\,k_1^2\right)^{-1}. \tag{27}$$

The spatially uniform solution of Eqs. (17)–(20) has then small seeds 3 and 4 growing exponentially with the rate

$$\gamma = V|b_1 b_2| \approx 3\,\omega_e^2 k_{1\perp}^2 |a_1|^2 \left(2\sqrt{\omega_3\omega_4}\,k_1^2\right)^{-1}. \tag{28}$$

For $a_1 = 0.1$, $k_{1\perp} = k_1/7$, $\omega_4 = \omega_1/5$, the rate is $\gamma \approx 4.7\times 10^{-4}\,\omega_e^2/\omega_1$. For the laser wavelength $\lambda_1 \approx 350\,\mathrm{nm}$, as at National Ignition Facility [3], and $\omega_e = \omega_1/50$, corresponding to the plasma concentration $n_0 \approx 3\times 10^{18}\,\mathrm{cm}^{-3}$, the distance within which the growing pulse amplitude makes one exponentiation is $c/\gamma \approx 30\,\mathrm{cm}$. To keep this distance the same at succeeding stages of the spectral energy transfer, occurring at shorter laser wavelengths, the plasma concentration should be increasing in proportion to the laser frequency. Then, the final stage of 10-stage cascade, converting micrometer laser wavelengths energy to nanometer wavelengths, would require the plasma concentration $n_0 \approx 3\times 10^{21}\,\mathrm{cm}^{-3}$. The first-stage plasma concentration can be produced by ionizing gas, the 10th-stage concentration can be produced by ionizing foam. Compared to Raman amplification of laser pulses in plasma [7], the plasma here is easier to implement, since it need not be homogeneous.

V. CONTROL OF NONLINEAR DETUNING

As the seed amplitude grows and the pump depletes, the initially perfect resonance may be detuned by nonlinear frequency shifts $\delta\omega_j$. These shifts do not exhibit automatic cancellation of the leading terms for paraxial pulses and thus can much exceed the coupling exhibiting such a cancellation. The detuning, $\delta\omega = \delta\omega_4 + \delta\omega_3 - \delta\omega_2 - \delta\omega_1$, can be controlled by using "dual seeds," coupled like pump pulses 1 and 2. Let all pulses have the same polarization, and let seed pulse 5 be close to 3 in amplitude $|b_5| \approx |b_3|$, and frequency $\omega_5 \gg \omega_3 - \omega_5 \gg \omega_e$, while $\boldsymbol{k}_{5\perp} \approx -\boldsymbol{k}_{3\perp}$. Pulse 6, resonantly amplified with pulse 5 by the same pumps 1 and 2, would then satisfy conditions $\omega_4 \gg \omega_6 - \omega_4 \gg \omega_e$, $\boldsymbol{k}_{6\perp} \approx -\boldsymbol{k}_{4\perp}$, $|b_6| \approx |b_4| \approx |b_3|$. The nonlinear detuning of each of the dual resonances would then be

$$\begin{aligned}\delta\omega \approx{}& \frac{\omega_e^2|b_4|^2}{2\omega_4^2}\left[\frac{(\boldsymbol{k}_6-\boldsymbol{k}_4)^2}{(k_6-k_4)^2}-3-\frac{2\omega_4}{\omega_3}+\frac{4\omega_4}{\omega_1}\right]+\frac{\omega_e^2|b_3|^2}{2\omega_3^2}\\ &\times\left[\frac{(\boldsymbol{k}_3-\boldsymbol{k}_5)^2}{(k_3-k_5)^2}-3-\frac{2\omega_3}{\omega_4}+\frac{4\omega_3}{\omega_1}\right]-\frac{\omega_e^2|b_1|^2}{\omega_1^2}\\ &\times\left[\frac{(\boldsymbol{k}_2-\boldsymbol{k}_1)^2}{(k_2-k_1)^2}-3+\frac{\omega_1}{\omega_4}+\frac{\omega_1}{\omega_3}\right].\end{aligned} \tag{29}$$

It can be zeroed out by proper selection of the ratios between the transverse and longitudinal components of vectors $\boldsymbol{k}_6 - \boldsymbol{k}_4 = \boldsymbol{k}_3 - \boldsymbol{k}_5$ and $\boldsymbol{k}_2 - \boldsymbol{k}_1$.

VI. PUMP DEPLETION REGIME FOR FULLY OVERLAPPING PULSES

At zeroed out resonance detuning, there is a simple analytical solution of Eqs. (17)–(20), where all b_j keep constant phases synchronized such that $\arg b_6 + \arg b_5 = \arg b_4 + \arg b_3 = \arg b_2 + \arg b_1 - \pi/2$, while the intensities are given by the formulas

$$\begin{aligned}&|b_3|^2 = |b_4|^2 = |b_5|^2 = |b_6|^2 \equiv I_{\mathrm{seed}}/4,\\ &|b_1|^2 = |b_2|^2 \equiv I_{\mathrm{pump}}/2, \quad I_{\mathrm{seed}} = I_{\mathrm{seed0}} + I_{\mathrm{pump0}} - I_{\mathrm{pump}}\\ &I_{\mathrm{pump}} = \frac{(I_{\mathrm{pump0}} + I_{\mathrm{seed0}})I_{\mathrm{pump0}}}{I_{\mathrm{pump0}} + I_{\mathrm{seed0}}\exp(2\gamma t)}.\end{aligned} \tag{30}$$

Within a few growth times γ^{-1}, nearly all the pump energy is converted into the seed pulses 3 and 5 of nearly doubled frequency. Equation (30) can be generalized for multiple pumps amplifying multiple seeds.

The spatially uniform solution is applicable when all pulses nearly completely overlap throughout the amplification process. The radius of a pump pulse of power P_1 is

$R \approx 2\sqrt{2P_1}c(a_1\omega_e\sqrt{P_{\rm cr}})^{-1}$. For $a_1 \approx 0.1$, $\lambda_1 \approx 350$ nm, $\omega_e = \omega_1/50$, and $P_1 \approx P_{\rm cr} \approx 40$ TW, it means $R \approx 0.08$ mm. The transverse slippage between such two pump pulses having opposite $\boldsymbol{k}_{2\perp} = -\boldsymbol{k}_{1\perp}$, $k_{1\perp} = k_1/7$, can be prevented by a grazing angle reflecting these pulses from mirrors forming a kind of channel. The longitudinal slippage between the pump and amplified pulses can be neglected for a large pulse lengths, say, 30 cm (corresponding to the duration 1 ns and each input pump energy 40 kJ). The amplified output pulses 3 and 5 could be used then as input pump pulses for the second stage of the laser energy transfer to shorter wavelengths, and so on. The spectral transfer of larger laser energies could be accomplished in multiple channels or by using multiple weakly coupled pulses in a larger radius channel.

VII. LONGITUDINAL PULSE-SLIPPAGE REGIME

Very short seed pulses can be amplified in regimes employing, rather than avoiding, longitudinal slippage between seed and pump pulses. Let a small seed pulse 3 enter at $t = 0$ the rear edge of fully overlapping pump pulses 1 and 2 having close frequencies and equal numbers of photons. The four-photon amplification occurs at $t > 0$ in the domain $c_1t < z < c_3t \leqslant c_1t + L$, where $c_j \approx c(1 - k_{j\perp}^2/2k_j^2)$ is the longitudinal group velocity of pulse j, and L is the pump length. At an advanced nonlinear stage, the amplified pulse amplitude is

$$|b_3| \approx |b_{10}|\sqrt{k_4/k_3}\,\{1 + \exp[2(\zeta_M - \zeta)]\}^{-1/2}\tau/\zeta_M,$$
$$\tau = 2\gamma t, \quad \zeta = 2\gamma\sqrt{t(c_3t - z)/(c - c_4)}, \tag{31}$$

$$\zeta_M \exp\zeta_M \approx \tau\sqrt{k_4/k_3}\,|b_{10}|/|b_{30}| \gg 1, \tag{32}$$

in the domain $\zeta > 0$, $\zeta - \zeta_M \ll \zeta_M$; in the overlapping domain $\zeta - \zeta_M \gg 1$, where pumps are already depleted,

$$|b_3| \approx |b_{10}|\sqrt{k_4/k_3}\,\tau\tilde{\zeta}_M/\zeta^2, \quad \tilde{\zeta}_M \approx \zeta_M + \ln(\zeta_M\tilde{\zeta}_M/\zeta^2). \tag{33}$$

The pulse amplitude and energy (located at $\zeta \sim \zeta_M$) grow approximately linearly with the amplification time, while the pulse duration decreases approximately inversely to the amplitude. For simplicity, we ignored the dual seed pulse in these formulas, but it can be taken into account like in the preceding section.

VIII. DISCUSSION

Our paper presents several important innovations which jointly accomplish a major conceptual breakthrough in the theory and applications of relativistic four-photon scattering in plasma.

First, we develop a very compact general description of the relativistic four-photon scattering, including noncollinear laser pulses which have not been studied adequately in the literature.

Second, we show that sufficiently noncollinear laser pulses can jointly propagate without self-focusing and transverse filamentation even at total powers exceeding the critical power of relativistic self-focusing. This is because such pulses, in contrast to collinear pulses, can be weakly coupled and stable at individual, rather than total powers, not exceeding the critical power. As seen from Fig. 1, multiple pairs of noncollinear laser pulses can have the same sum of wave vectors $\mathbf{K}$ and frequencies 2ω and thus be in the same four-photon resonance. This allows, in principle, simultaneous amplification of multiple laser seeds by multiple laser pumps, even at total powers exceeding the critical power of relativistic self-focusing.

Third, we calculate the growth rate of four-photon amplification and show that, at mildly relativistic intensities, it can be sufficient for accomplishing the amplification within reasonable distances.

Fourth, we show how to stay within the four-photon resonance throughout the entire amplification process, despite varying nonlinear frequency shifts exceeding considerably the resonance bandwidth. This is achieved by using novel "dual seeds," which secure mutual cancellation of the frequency shifts in the synchronism conditions. An incidental benefit is that unwanted resonances may not survive.

Fifth, we show how to prevent the energy flow reversal back from intense amplified pulses to pumps. This is achieved by using pumps with the same number of photons. Such pumps are depleted simultaneously, which ensures nearly total energy transfer. Any small leftover of pump energy, due to inexact matching of numbers of photons, just slightly reduces the efficiency. The process can realistically be terminated for multiple pump pulses simultaneously, before any reversal of the energy flow occurs. This is because the rate of energy transfer drops significantly when pump amplitudes become small. For example, if the pump leftover energy is 10% of the initial pump energy, the distance within which pumps stay that small is 10 times larger than the initial pump depletion distance, which leaves an ample space for terminating the interaction. Incidentally, these means of preventing the energy flow reversal might suggest tools for controlling more general inverse cascades. In particular, this might help improving the kinetic method of suppressing the relativistic filamentation instability by phase randomization of powerful laser pulses [30,31], impeded by the well-known tendency of Bose-Einstein condensate formation via the inverse energy cascade [32–34].

Sixth, in the four-photon amplification of noncollinear laser pulses, the transverse slippage of the pulses could be an issue. It can be prevented by side mirrors reflecting pulses at grazing angles as needed. For pulses fully overlapped in the transverse directions, we present simple analytical solutions of the evolution equations, showing energy transfer to a nearly double-frequency seed up to the total pump depletion, addressing both the cases of negligible or substantial longitudinal slippages.

Notably, in our scheme, plasma need not be too homogeneous. We propose to use a plasma of very undercritical density, where the electron plasma frequency is much smaller than the laser frequencies, so that the four-photon resonance synchronism conditions are basically the same as in vacuum, unaffected by plasma inhomogeneities. At the same time, the very same plasma inhomogeneities may, in fact, serve to suppress parasitic processes such as Raman or Brillouin scattering, mediated by plasma waves or sound waves, which are much more sensitive to inhomogeneities than the photons that we consider [35,36]. The parasitic processes involving plasma waves can be further suppressed by laser frequency chirping [37], without affecting the useful four-photon resonance.

Although the spectral transfer of optical energy to x-ray energy is, in principle, very efficient, the achievable experimental efficiencies might be less in practice for reasons unanticipated here. If each of the 10 stages were, say, only 50% efficient, then a factor of as much as 1000 would be lost in the 10 stages required through frequency doubling, resulting in kilojoules in the x-ray regime rather than megajoules. However, considering that at present only millijoules x rays are available, even attaining kilojoules would be a fantastic advance. While we do not emphasize here applications, it can be expected that short-wavelength high-energy laser pulses will enable radically new discoveries and technologies. An example of new technologies that might be readily imagined is the delivery of laser power to the compressed target core for achieving fast ignition in inertial confinement fusion, not with lasers in the optical range [38], but with even kilojoules at x-ray wavelengths capable of naturally penetrating even very dense plasma layers.

In summary, the proposal advanced here is unique in its ambition. It identifies the methodologies that can be used for a highly efficient resonant plasma-based spectral transfer of huge energies of short optical laser pulses to deep-ultraviolet and x-ray wavelengths. Apart from the evident importance of this for applications, the methodologies put forth here are of basic academic significance.

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