Super-resonant four-photon collinear laser frequency multiplication in plasma

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Resonant four-photon scattering could nearly double frequencies of intense laser pulses in plasma. However, transverse slippage between pulses presents a technological challenge, while collinear four-photon scattering is forbidden for classical light dispersion in plasma. Nonlinear renormalization of intense laser pulses can enable collinear four-photon resonance. However, such a very intensity-sensitive resonance is difficult to maintain for evolving pulses. Remarkably, there is a lower-dimensionality submanifold of the resonant four-photon manifold where the evolving pulses stay in resonance. This could enable an all-optical frequency doubling of mildly relativistic-intense laser pulses in collinear geometry, advantageously free of the transverse slippage challenges.

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I. INTRODUCTION

The lack of a material that can reflect laser pulses of energy fluences greater than a few J/cm² limits practically achievable pulse energies. Due to this limitation, hundreds of square meters of high quality mirrors for laser resonant cavities and reflecting gratings for the chirped pulse amplification technique [1,2] are needed to produce megajoule laser pulse energies at the National Ignition Facility (NIF) [3–7] or Laser Mégajoule Project [8–10].

Additionally, the lack of a material that can reflect photons of energies greater than 10 eV, corresponding to wavelengths shorter than $1/8 \mu m$, limits direct production of laser pulses at such wavelengths to a single pass amplification. As a result, the currently achievable laser pulse energies sharply decrease from megajoules at optical wavelengths to millijoules at substantially submicron wavelengths. Such millijoule ultraviolet and x-ray pulses are now produced by free electron lasers [11,12].

It would be of a great interest to find an efficient way of transferring megajoule laser energies from the optical to significantly shorter wavelengths. As but one possible application, in inertial confinement fusion, an extremely large frequency up-shift would enable the direct delivery of laser pulse energy to the compressed target core, producing the fast ignition of a small fraction of fuel, which would be a major step towards the ultimate NIF goal.

Even a moderately large frequency up-shift could be very useful for the NIF. The NIF has 192 laser pulses, 10-kJ energy each, at 351-nm wavelength. These pulses are produced by 50% efficient frequency tripling of 1053-nm laser pulses. The tripling is accomplished in a standard nonlinear material in two stages. In the first stage, the frequency is doubled. In the second stage, the original input frequency is added to the double frequency output of the first stage. It would be beneficial to avoid this very large and expensive frequency tripling system, but the higher frequency is needed for more robust propagation of laser pulses in plasma. Large frequency up-shifts could potentially be achieved via resonant nonlinear wave interactions in plasma. However, propagation of intense laser pulses in plasma tends to be disrupted by various instabilities. The instabilities could be mitigated at large laser-to-plasma frequency ratios, but low-frequency plasma waves cannot resonantly produce significant shifts of the laser pulse frequency.

It was proposed to use the resonant four-photon scattering in rarefied plasma for transferring megajoule laser energies from optical to shorter wavelengths [13]. In the regimes [13], the scattering rates were proportional to the square of angles between the laser pulses, and, to accomplish energy transfers within modest laboratory distances, these paraxial angles should not be too small. However, moderately small paraxial angles already present a technological challenge by producing significant transverse slippages between the pulses.

Collinear four-photon scattering would be advantageously free of the transverse slippage challenges. Additionally, collinear regimes would benefit from the transverse slippage suppressing parasitic noncollinear scattering seeded by noise. Collinear pulses could conveniently propagate in a shallow plasma channel, in the ground-state transverse mode, limiting the transverse size of the pulses. The pulse power could be kept below the threshold of the self-focusing instability [14–17], thus excluding this kind of impediment as well.

While the classical light dispersion in plasma does not allow resonant collinear four-photon scattering, such scattering can be enabled by nonlinear renormalization of laser frequencies and be fast enough to occur within modest laboratory distances [18]. However, it would be challenging to maintain the resonance as the interaction proceeds and the laser intensities evolve. This is because the changes in laser intensities would typically cause the renormalized laser frequencies to change by more than the width of four-photon resonance. Therefore, any prearranged exactly resonant conditions would typically be ruined quickly by detuning of the resonance in the process of natural evolution. Our goal here is to try to find a submanifold of the resonant manifold in parameter space, such that the evolution starting in this submanifold proceeds within it. This hypothetical submanifold is called here "super-resonant."

To facilitate reading of the paper, we briefly outline its content. In Sec. II, using a generic form of four-wave evolution equations in canonical variables [19], we specify, in general terms, the regimes to be examined. In Sec. III, we derive particular equations for renormalized frequencies and slowly varying envelopes of copropagating plane laser wave packets of the same linear polarization. In Sec. IV, we simplify these equations for the most interesting regimes, where the laser frequency is nearly doubled. In Sec. V, we find explicit examples of super-resonant solutions in such frequency-doubling regimes. In Sec. VI, we summarize and briefly discuss the results.

II. REGIMES TO BE EXAMINED

The synchronism conditions needed for exactly resonant scattering of two plane collinear waves into two plane collinear waves have the form

$$k_1 + k_2 = k_3 + k_4 \equiv 2k, \quad \omega_1 + \omega_2 = \omega_3 + \omega_4 \equiv 2\omega, \quad (1)$$

where k_j are wave numbers and ω_j are frequencies of the waves. As long as the exact resonance is maintained, the evolution equations for properly defined wave envelopes b_j can be presented in the following canonical form:

$$\iota(\partial_t + c_1 \partial_z)b_1 = V b_2^* b_3 b_4, \quad \iota(\partial_t + c_2 \partial_z)b_2 = V b_1^* b_3 b_4, \quad (2)$$

$$\iota(\partial_t + c_3 \partial_z) b_3 = V^* b_1 b_2 b_4^*, \quad \iota(\partial_t + c_4 \partial_z) b_4 = V^* b_1 b_2 b_3^*, \quad (3)$$

where c_j are the group velocities of waves. The same onedimensional equations (with a slightly modified coupling coefficient V) are, in fact, applicable for pulses in a shallow channel ground-state transverse mode.

The standard four-wave scattering rate is quadratic in wave amplitudes, as are the nonlinear corrections to the wave frequencies. Though coming from the same cubic nonlinearity, these quantities are not necessarily of the same magnitude. For noncollinear four-photon scattering regimes [13], the scattering rate is typically much smaller than the nonlinear corrections to photon frequencies, because of the mutual cancellation of leading physical contributions to the rate. This makes keeping the resonance between the evolving pulses challenging. The nonlinear corrections to the photon frequencies might be canceled by arranging multiple noncollinear seed pulses [13], but it is difficult to manage simultaneously the transverse slippage between the pulses. The transverse slippage could be managed by a grazing angle reflection of the pulses from outside mirrors [13]. These mirrors could also provide resonant synchronism between the pulses over repeated meetings in the plasma. However, this solution is technologically difficult.

Highly desirable would be regimes where the collinear four-photon resonance [18] persists within the plasma, absent transverse slippage between the pulses. However, the challenge in finding such collinear regimes is compounded by the collinear four-photon resonance (created rather than only adjusted by the renormalization) being much more sensitive to intensity variations than the noncollinear four-photon resonance. It is not at all clear at the outset that the hypothetical super-resonant submanifold, where evolving collinear pulses stay in the four-photon resonance, exists. Being of a lower, if any, dimensionality, the super-resonant submanifold hardly could be found numerically without detailed analytical guidance. (Note that a similar difficulty was illustrated in a preliminary search of the enduring resonant regimes for noncollinear four-photon scattering where empirical numerical approaches fell far short of finding the theoretically possible energy transfers, even when using a simplified model that disregarded the transverse slippage between pulses [20].) Hence, what is critically needed is to examine analytically if such hard-to-find super-resonant regimes, where evolving collinear pulses stay in enduring four-photon resonance, are possible.

Of our highest interest here are the scattering regimes where the output amplified seed pulse 3 has nearly double the wave number of the mean input pump wave number:

$$k_4 = 2k - k_3 \ll k.$$
 (4)

Existence of such a collinear four-photon resonance for small seeds was shown in [18]. It still needs to be verified for larger seeds 3 and 4, as a prerequisite for searching the super-resonant regimes.

In regimes (4), nearly all the pump energy goes into the short-wavelength amplified pulse 3 and just a small fraction of the energy goes into the long-wavelength disposable pulse 4. This may be possible at laser frequencies much greater than the plasma frequency:

$$\omega_j \gg \omega_e = \sqrt{4\pi n_0 e^2/m} \equiv k_e c. \tag{5}$$

Here *m* is the electron rest mass, -e is the electron charge, n_0 is the electron concentration of plasma in absence of lasers, and *c* is the speed of light in vacuum.

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Laser group velocities c_j are then close to c and not sensitive to nonlinear corrections which can be therefore neglected in c_j , so that

$$c_i = c - \delta c_i, \quad \delta c_i \approx c k_e^2 / 2k_i^2. \tag{6}$$

The speed of longitudinal slippage of the disposable pulse 4 is much greater than between other pulses.

Consider regimes in which the slippage between pulses 1, 2, and 3 can be neglected, as depicted in Fig. 1. Then, in the variables t, $\zeta = z - c_1 t$, Eqs. (2) and (3) reduce to

$$\iota \partial_t b_1 \approx V b_2^* b_3 b_4, \quad \iota \partial_t b_2 \approx V b_1^* b_3 b_4, \tag{7}$$

$$\iota \partial_t b_3 \approx V^* b_1 b_2 b_4^*, \quad \iota (\partial_t - \delta c_4 \partial_\zeta) b_4 \approx V^* b_1 b_2 b_3^*, \qquad (8)$$

where the partial derivative ∂_t is already taken at fixed ζ rather than *z*. Solutions of these equations satisfy the following Manley-Rowe relations:

$$\tilde{N} \equiv |b_3|^2 - |b_{30}|^2 \approx |b_{10}|^2 - |b_1|^2 \approx |b_{20}|^2 - |b_2|^2, \quad (9)$$

where additional indices "0" are used for initial conditions. The physical meaning of these relations is that the variation of photon concentration \tilde{N} in pulse 3 has the opposite sign and the same value as in each of the pump pulses 1 and 2. We'll consider fully overlapping pulses 1, 2, and 3 of the same length L_1 and flat photon concentrations within this length.



FIG. 1. The longitudinal slippage between pump pulses 1 and 2 and seed pulse 3 of nearly double pump frequency is negligible, so that positions of these three pulses do not change in the window moving with their joint group velocity. These pulses, ideally fully overlapping, are shown here exaggeratedly displaced to make more visible that they are three different pulses rather a single pulse. Also for better visibility, the aperture of all pulses is shown here exaggeratedly large (while, in fact, it is small due to the mildly relativistic intensities at powers below the self-focusing power). The low frequency disposable seed pulse 4 has a smaller group velocity, so that it slips backwards in this moving window.

For nearly simultaneous depletion, the initial photon concentrations in pump pulses need to be close: $|b_{10}|^2 \approx |b_{20}|^2$.

If even the slippage of pulse 4 over pulses 1-3 were negligible within the propagation length *L*,

$$\delta L_4 \approx L \delta c_4 / c \approx L k_e^2 / 2k_4^2 \ll L_1, \tag{10}$$

pulse 4, overlapping with pulses 1–3, would also satisfy the Manley-Rowe relation:

$$|b_4|^2 - |b_{40}|^2 \approx \tilde{N}.$$
 (11)

Then, assuming that the wave numbers k_j and renormalized frequencies

$$\omega_j \approx ck_j + ck_e^2/2k_j + \sum_l V_{j,l}|b_l|^2$$
(12)

stay in the exact resonance (1) until the pump pulses are significantly depleted, the photon concentration in disposable pulse $|b_4|^2$ would ultimately increase to about $|b_{10}|^2 \sim |b_{20}|^2$. The respective increase of the photon energy density $|b_4|^2\omega_4$ in pulse 4 would be about $|b_{10}|^2\omega_4$. In the terms of electron quiver velocities v_j , the energy density $|b_j|^2\omega_j$ is proportional to $v_j^2\omega_j^2$, so that $v_j^2 \propto |b_j|^2/\omega_j \propto |b_j|^2/k_j$. It follows that v_4^2 would ultimately increase to about v_{10}^2k/k_4 . For smallness of higher order nonlinearities, electron quiver velocities v_j should be much smaller than the speed of light c:

$$a_j = v_j/c \ll 1. \tag{13}$$

To keep $a_4 \ll 1$, it would be needed then to use excessively small input pump intensities:

$$a_{10}^2 \sim a_{20}^2 \ll k_4/k. \tag{14}$$

This would, in turn, imply excessively large amplification distances. Such excesses could be avoided by disposing pulse

4 to a reasonable level $a_4 \sim a_{10}$ allowing us to preserve the natural limitation (13) for pump pulses. The disposing could occur in short regions of a denser plasma significantly slowing down pulse 4 while virtually not affecting the higher frequency pulses 1–3. Within such a region, pulse 4 could be quickly replaced by its fresh version of a slightly smaller amplitude. The pulse 4 energy disposed at $a_4 \sim a_{10}$ is just a small $k_4^2/k^2 \ll 1$ fraction of the input pump energy. Therefore, any number of such disposals, much smaller than $k^2/k_4^2 \gg 1$, would not noticeable deteriorate high efficiency of the pulse 3 amplification.

A continual disposing could be accomplished in the regimes where the slippage of disposable pulse 4 is large $\delta L_4 \gg L_1$ and roughly the same as the pulse 4 length $L_4 \sim \delta L_4$. The input ratio of the pulse 4 energy to the pump energy is small:

$$\frac{k_4|b_{40}|^2\delta L_4}{(k_1|b_{10}|^2+k_2|b_{20}|^2)L_1} \sim \frac{a_{40}^2}{2a_{10}^2}\frac{\delta L_1}{L_1} \ll 1,$$
(15)

for $a_{40} \leq a_{10}$ and $\delta L_1 \ll L_1$. Rear edges of all four pulses coincide at t = 0. Then, pulse 4 slips backwards over pulses 1–3 with the speed δc_4 . Each layer of pulse 4 slips over the common length L_1 of pulses 1–3 quickly, so that a noticeable resonance detuning does not occur on such a short timescale. Thus, pulses 1–3 effectively interact at each time with a fresh layer of pulse 4. To keep the resonance throughout the evolution, each layer of pulse 4 needs to be prearranged to get into resonance with pulses 1–3 upon encountering them. The layer phase also needs to be prearranged properly.

III. BASIC EQUATIONS

We start with the nonlinear evolution equation [13] for dimensionless vector potential $\vec{a} = e\vec{A}/mc^2$ of the electromagnetic field, derived by expansion in $a \ll 1$ (meaning that the electron quiver velocity in the laser field is much smaller than the speed of light in vacuum *c*). For plane waves propagating along the axis *z* and polarized along the axis *x*, so that $\vec{a} = a\vec{e_x}$, the evolution equation takes the form [18]

$$(\partial_t^2 - c^2 \partial_z^2 + \omega_e^2)a$$

= $\omega_e^2 a [1 - (\partial_t^2 + \omega_e^2)^{-1} c^2 \partial_{zz}] a^2 / 2 + O(a^5).$ (16)

Solutions of the equation (16) are searched in the form

$$a = \sum_{j} (a_j e^{i\phi_j} + \text{c.c.}) + \delta a, \ \partial_z \phi_j = k_j, \ \partial_t \phi_j = -\omega_j, \quad (17)$$

where envelopes a_j slowly vary in space-time while δa represents small nonresonant beatings generated by nonlinearity.

The renormalized relation between ω_j and k_j is defined by matching all terms proportional to a_j , which gives, neglecting higher order terms,

$$\omega_j^2 = c^2 k_j^2 + \omega_e^2 + \omega_e^2 \left(|a_j|^2 F_{j,j}/2 + \sum_{l \neq j} |a_l|^2 F_{j,l} \right), \quad (18)$$

$$F_{j,l} = \frac{c^2 (k_j - k_l)^2}{(\omega_j - \omega_l)^2 - \omega_e^2} + \frac{c^2 (k_j + k_l)^2}{(\omega_j + \omega_l)^2 - \omega_e^2} - 3 = F_{l,j}.$$
 (19)

The evolution equations for slowly varying envelopes a_j can be derived from (16). Neglecting the second derivatives of a_j 's, the envelopes just move with the wave group velocities and can additionally slowly vary due to non-linear interactions. We are interested here in the regimes where four-photon scattering is not coupled with the Raman scattering. This implies that the beatings of input waves are well off the Raman resonances corresponding to zeros of denominators $(\omega_j - \omega_l)^2 - \omega_e^2$ in (19). Assuming that the parasitic Raman scattering seeded by noise is suppressed, the nonlinear evolution of wave envelopes, caused by four-photon scattering, can be described by the following equations:

$$2\iota(\omega_{j}\partial_{t} + c^{2}k_{j}\partial_{z})a_{j} = \omega_{e}^{2}\sum_{\substack{p\neq j, s\neq j \\ p\neq j, s\neq j}}a_{l}^{*}a_{p}a_{s}F_{l;p,s}F_{j,l;p,s}, \quad (20)$$

$$F_{l;p,s} = F_{l;s,p} = \frac{c^{2}(k_{p} - k_{l})^{2}}{(\omega_{p} - \omega_{l})^{2} - \omega_{e}^{2}}$$

$$+ \frac{c^{2}(k_{s} - k_{l})^{2}}{(\omega_{s} - \omega_{l})^{2} - \omega_{e}^{2}} + \frac{c^{2}(k_{p} + k_{s})^{2}}{(\omega_{p} + \omega_{s})^{2} - \omega_{e}^{2}} - 3, \quad (21)$$

$$F_{j,l;p,s} = \langle e^{\iota(\phi_p + \phi_s - \phi_l - \phi_j)} \rangle = F_{l,j;p,s} = F_{j,l;s,p} = F_{p,s;j,l}^*.$$
(22)

Here, the angle brackets signify averaging over a space-time domain small compared to the space-time scales of envelopes variation, but large compared to the scales of nonresonant beatings variation.

In the exact resonance (1), factors $F_{l;p,s}$, defined by (21), have additional symmetries,

$$F_{2;3,4} = F_{1;3,4} = F_{3;1,2} = F_{4;1,2},$$
(23)

while factors $F_{i,l;p,s}$ can be reduced to

$$F_{1,2;3,4} = F_{3,4;1,2} = 1 \tag{24}$$

by proper selection of integration constants in phases ϕ_j (17). Then, Eq. (20) takes, in the variables t, ζ , b_j ,

$$a_i(t,z) = b_i(t,\zeta)/\sqrt{k_i},$$
(25)

the form of Eqs. (7) and (8) with

$$V = \frac{ck_e^2 F_{2;3,4}}{\sqrt{k_1 k_2 k_3 k_4}}.$$
 (26)

Equation (18) reduces to Eq. (12) with

$$V_{j,j} = \frac{ck_e^2}{4k_j^2} F_{j,j}, \quad V_{j,l\neq j} = \frac{ck_e^2}{2k_l^2} F_{j,l}.$$
 (27)

IV. LARGE FREQUENCY UPSHIFTS

For laser frequencies much greater than the plasma frequency (5), coefficients $F_{j,j}$ (19) reduce to

$$F_{j,j} \approx -2.$$
 (28)

Well off the Raman resonances $(\omega_j - \omega_l)^2 - \omega_e^2 = 0$, coefficients $F_{j,l}$ (19) reduce to

$$F_{j,l} \approx \frac{k_e^2}{(k_j - k_l)^2 - k_e^2} - 1.$$
 (29)

Taking into account that $k_4 \ll k$ (4), $F_{j,l\neq j}$ can be further simplified to

$$F_{1,3} \approx F_{1,4} \approx F_{2,3} \approx F_{2,4} \approx F_{3,4} \approx -1,$$
 (30)

$$F_{1,2} \approx \frac{k_e^2}{(k_1 - k_2)^2 - k_e^2} - 1,$$
(31)

while the factor $F_{2;3,4}$ (21) reduces to

$$F_{2;3,4} \approx k_e^2 \left(\frac{1}{k_1^2} + \frac{1}{k_2^2} + \frac{1}{4k^2} \right).$$
(32)

The resonance condition (1) for renormalized frequencies (12) can be presented in the form

$$\binom{k_1k_2}{k_3k_4} - 1 (1 - |a_1|^2 - |a_2|^2 - |a_3|^2 - |a_4|^2)$$

$$\approx \frac{k_e^2(k_1|a_1|^2 + k_2|a_2|^2)}{k[(k_1 - k_2)^2 - k_e^2]}.$$
(33)

Taking into account that $k_4 \ll k$ (4) and $|a_j| \ll 1$, it follows that

$$(k_1 - k_2)^2 - k_e^2 \approx 2k_e^2 (|a_1|^2 + |a_2|^2)k_4/k.$$
(34)

This should be well off the Raman resonance, the frequency width of which is about $\omega_e^2 |a_1| / \omega_1$ [21,22]. That means

$$(k_1 - k_2)^2 - k_e^2 \gg 2k_e^3 |a_1|/k \implies 2|a_1|k_4 \gg k_e.$$
(35)

Within the resonant manifold (34), the factor $F_{2;3,4}$ (32) reduces to

$$F_{2;3,4} \approx \frac{9k_e^2}{4k^2}.$$
 (36)

V. SUPER-RESONANT REGIMES

The most promising are the regimes where pump pulses 1 and 2 jointly move together with seed pulse 3 which is at nearly twice their frequency. These three pulses, moving together, encounter fresh layers of the lower frequency longer disposable seed pulse 4 which slips backwards through pulses 1–3. Parameters of the plasma and pulse 4 at every encounter space-time location need to be prearranged to satisfy the resonance conditions (1) and phase synchronism condition:

$$\arg b_4 = \arg b_1 + \arg b_2 - \arg b_3 - \pi/2.$$
 (37)

These may leave some flexibility in choosing parameters, as seen from the simplified equation (34) for the resonant manifold. The flexibility may be employed to facilitate the useful amplification and suppression of parasitic instabilities. However, our goal here is just to verify the existence of superresonant regimes in the parameter domain of physical interest. Therefore, we will choose values of flexible parameters in an easy to illustrate though not necessarily optimal way.

For a small initial amplitude of seed pulse 3, the Manley-Rowe relations (9) reduce to

$$|b_1|^2 \approx |b_{10}|^2 - |b_3|^2, \quad |b_2|^2 \approx |b_{20}|^2 - |b_3|^2.$$
 (38)

For simultaneously depleting pump pulses 1 and 2, their initial photon concentrations need to be the same:

$$|b_{20}|^2 = |b_{10}|^2 \equiv b_0^2.$$
(39)

Then, the pulse 3 evolution equation (8) reduces to

$$\partial_t |b_3| \approx V |b_4| (b_0^2 - |b_3|^2),$$
(40)

with the factor V, according to (26) and (36), given by

$$V \approx \frac{9ck_e^4}{4k^3\sqrt{2kk_4}}.$$
(41)

If even the slippage of pulse 4 were negligible, the additional Manley-Rowe relation (11) would be valid, implying $|b_4| \approx |b_3|$. The respective solution of the equation (40) would be

$$|b_1|^2 \approx b_0^2 - |b_3|^2 \approx \frac{b_0^2}{1 + \exp\left(2Vb_0^2t\right)|b_{40}|^2/b_0^2}.$$
 (42)

The propagation distance for nearly complete depletion of pump pulses, leaving not yet depleted just a small fraction $\alpha \ll 1$ of the input pump power, would be

$$L_d \approx \frac{c}{2Vb_0^2} \ln\left(\frac{b_0^2}{\alpha |b_{40}|^2}\right). \tag{43}$$

This could be rewritten in the form

$$L_d \approx \frac{\lambda}{9\pi |a_{10}|^2} \frac{k^4}{k_e^4} \sqrt{\frac{2k_4}{k}} \ln\left(\frac{k|a_{10}|^2}{\alpha k_4 |a_{40}|^2}\right), \quad (44)$$

where $\lambda \approx 2\pi/k$ is the laser pump wavelength, and $|a_{10}|^2 \ll k_4/k$ (14).

For the regimes where slippages of pulse 4 are large, there is a flexibility in choosing $|b_4|$. Let us, for example, choose it to be constant, $|b_4| = |a_{40}|\sqrt{k_4}$. The respective solution of the equation (40) is

$$b_0 - |b_3| \approx \frac{2b_0}{1 + \exp(2Vb_0|b_4|t)}.$$
 (45)

The propagation distance for nearly complete depletion of pump pulses is

$$L_d \approx \frac{c}{2Vb_0|b_4|} \ln\left(\frac{4}{\alpha}\right) \approx \frac{\lambda\sqrt{2}}{9\pi |a_{10}||a_{40}|} \frac{k^4}{k_e^4} \ln\left(\frac{4}{\alpha}\right).$$
(46)

It can be shorter than (44), due to the softer limitations on initial amplitudes, $|a_{10}| \ll 1$ and $|a_{40}| \ll 1$.

- [1] D. Strickland and G. Mourou, Compression of amplified chirped optical pulses, Opt. Commun. **55**, 447 (1985).
- [2] G. A. Mourou, C. P. J. Barty, and M. D. Perry, Ultrahigh Intensity Lasers: Physics of the Extreme on a Tabletop, Phys. Today 51(1), 22 (1998).
- [3] P. J. Wegner, J. M. Auerbach, J. Biesiada, A. Thomas, S. N. Dixit, J. K. Lawson, J. A. Menapace, T. G. Parham, D. W. Swift, P. K. Whitman, and W. H. Williams, NIF final optics system: Frequency conversion and beam conditioning, in *Optical Engineering at the Lawrence Livermore National Laboratory II: The National Ignition Facility*, SPIE Conf. Proc. No. 5341, edited by M. A. Lane and C. R. Wuest (SPIE, Bellingham, WA, 2004), p. 180.
- [4] M. L. Spaeth, K. R. Manes, D. H. Kalantar, P. E. Miller, J. E. Heebner, E. S. Bliss, D. R. Spec, T. G. Parham, P. K. Whitman,

For a numerical example with $\alpha = 0.1$, corresponding to 90% pump depletion, $k = 20k_e$, and $|a_{40}|^2 = |a_{10}|^2 = 1/5$, (46) gives

$$L_d \approx 1.5 \times 10^5 \,\lambda. \tag{47}$$

For $\lambda = 3 \times 10^{-5}$ cm, this depletion length is $L_d \approx 4.5$ cm.

VI. SUMMARY

In contrast to the classical case, the four-photon resonance explored here is created (rather than just slightly modified) by nonlinearity. This makes the resonance very intensity sensitive and, generally, quickly detuned by the natural wave evolution. Remarkably, it appears nevertheless that there may exist a lower-dimensionality submanifold of the resonant four-photon manifold where evolution laser pulses stay in resonance. We derived a simple formula for such a superresonant submanifold in the most interesting regimes where the input pump energy is nearly completely transformed into the energy of an output pulse of nearly twice the pump frequency.

A few centimeter laser energy transfer distances in fully resonant regimes of our paper indicate that the transfer distances could stay submeter even if the resonance were occurring only in a few percentages of the laser propagation length. This would substantially relieve tough requirements to the plasma homogeneity by making acceptable random inhomogeneities which typically detune the four-photon resonance even ten times more than its width. Additionally, the respective detuning of the Raman resonance, greater by the factor of the laser-to-plasma square-frequency ratio, would facilitate suppression of parasitic Raman scattering. These regimes also promise softer requirements to the prearranged laser seeds and pumps, thus strongly motivating further study.

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P. J. Wegner, P. A. Baisden, J. A. Menapace, M. W. Bowers, S. J. Cohen, T. I. Suratwala, J. M. D. Nicola, M. A. Newton, J. J. Adams, J. B. Trenholme, R. G. Finucane, R. E. Bonanno, D. C. Rardin, P. A. Arnold, S. N. Dixit, G. V. Erbert, A. C. Erlandson, J. E. Fair, E. Feigenbaum, W. H. Gourdin, R. A. Hawley, J. Honig, R. K. House, K. S. Jancaitis, K. N. LaFortune, D. W. Larson, B. J. L. Galloudec, J. D. Lindl, B. J. MacGowan, C. D. Marshall, K. P. McCandless, R. W. McCracken, R. C. Montesanti, E. I. Moses, M. C. Nostrand, J. A. Pryatel, V. S. Roberts, S. B. Rodriguez, A. W. Rowe, R. A. Sacks, J. T. Salmon, M. J. Shaw, S. Sommer, C. J. Stolz, G. L. Tietbohl, C. C. Widmayer, and R. Zacharias, Description of the NIF Laser, Fusion Sci. Technol. **69**, 25 (2016).

[5] S. Le Pape, L. F. Berzak Hopkins, L. Divol, A. Pak, E. L. Dewald, S. Bhandarkar, L. R. Bennedetti, T. Bunn, J. Biener, J. Crippen, D. Casey, D. Edgell, D. N. Fittinghoff, M. Gatu-Johnson, C. Goyon, S. Haan, R. Hatarik, M. Havre, D. D.-M. Ho, N. Izumi, J. Jaquez, S. F. Khan, G. A. Kyrala, T. Ma, A. J. Mackinnon, A. G. MacPhee, B. J. MacGowan, N. B. Meezan, J. Milovich, M. Millot, P. Michel, S. R. Nagel, A. Nikroo, P. Patel, J. Ralph, J. S. Ross, N. G. Rice, D. Strozzi, M. Stadermann, P. Volegov, C. Yeamans, C. Weber, C. Wild, D. Callahan, and O. A. Hurricane, Fusion Energy Output Greater than the Kinetic Energy of an Imploding Shell at the National Ignition Facility, Phys. Rev. Lett. **120**, 245003 (2018).

- [6] A. L. Kritcher, A. B. Zylstra, D. A. Callahan, O. A. Hurricane, C. Weber, J. Ralph, D. T. Casey, A. Pak, K. Baker, B. Bachmann, S. Bhandarkar, J. Biener, R. Bionta, T. Braun, M. Bruhn, C. Choate, D. Clark, J. M. Di Nicola, L. Divol, T. Doeppner, V. Geppert-Kleinrath, S. Haan, J. Heebner, V. Hernandez, D. Hinkel, M. Hohenberger, H. Huang, C. Kong, S. Le Pape, D. Mariscal, E. Marley, L. Masse, K. D. Meaney, M. Millot, A. Moore, K. Newman, A. Nikroo, P. Patel, L. Pelz, N. Rice, H. Robey, J. S. Ross, M. Rubery, J. Salmonson, D. Schlossberg, S. Sepke, K. Sequoia, M. Stadermann, D. Strozzi, R. Tommasini, P. Volegov, C. Wild, S. Yang, C. Young, M. J. Edwards, O. Landen, R. Town, and M. Herrmann, Achieving record hot spot energies with large HDC implosions on NIF in HYBRID-E, Phys. Plasmas 28, 072706 (2021).
- [7] J. Tollefson *et al.*, US achieves laser-fusion record: What it means for nuclear-weapons research, Nature (London) 597, 163 (2021).
- [8] N. Fleurot, C. Cavailler, and J. Bourgade, The Laser Mégajoule (LMJ) Project dedicated to inertial confinement fusion: Development and construction status, Fusion Eng. Des. 74, 147 (2005).
- [9] J.-L. Miquel and E. Prene, LMJ & PETAL status and program overview, Nucl. Fusion 59, 032005 (2018).
- [10] V. Denis, M. Nicolaizeau, J. Néauport, C. Lacombe, and P. Fourtillan, LMJ 2021 facility status, in *Proceedings of SPIE* (SPIE, Bellingham, WA, 2021), Vol. 11666, p. 1166 603.
- [11] C. Pellegrini, A. Marinelli, and S. Reiche, The physics of x-ray free-electron lasers, Rev. Mod. Phys. 88, 015006 (2016).

- [12] J. Duris, S. Li, T. Driver, E. G. Champenois, J. P. MacArthur, A. A. Lutman, Z. Zhang, P. Rosenberger, J. W. Aldrich, R. Coffee, G. Coslovich, F.-J. Decker, J. M. Glownia, G. Hartmann, W. Helml, A. Kamalov, J. Knurr, J. Krzywinski, M.-F. Lin, J. P. Marangos, M. Nantel, A. Natan, J. T. O'Neal, N. Shivaram, P. Walter, A. L. Wang, J. J. Welch, T. J. A. Wolf, J. Z. Xu, M. F. Kling, P. H. Bucksbaum, A. Zholents, Z. Huang, J. P. Cryan, and A. Marinelli, Tunable isolated attosecond X-ray pulses with gigawatt peak power from a free-electron laser, Nat. Photonics 14, 30 (2020).
- [13] V. M. Malkin and N. J. Fisch, Towards megajoule x-ray lasers via relativistic four-photon cascade in plasma, Phys. Rev. E 101, 023211 (2020).
- [14] A. G. Litvak, Finite-amplitude wave beams in a magnetoactive plasma, Zh. Eksp. Teor. Fiz. **57**, 629 (1969); [Sov. Phys. JETP **30**, 344 (1970)].
- [15] C. E. Max, J. Arons, and A. B. Langdon, Self-modulation and self-focusing of electromagnetic waves in plasmas, Phys. Rev. Lett. 33, 209 (1974).
- [16] G.-Z. Sun, E. Ott, Y. C. Lee, and P. Guzdar, Self-focusing of short intense pulses in plasmas, Phys. Fluids 30, 526 (1987).
- [17] V. M. Malkin, On the analytical theory for stationary selffocusing of radiation, Physica D 64, 251 (1993).
- [18] V. M. Malkin and N. J. Fisch, Resonant four-photon scattering of collinear laser pulses in plasma, Phys. Rev. E 102, 063207 (2020).
- [19] V. E. Zakharov and E. A. Kuznetsov, Hamiltonian formalism for nonlinear waves, Usp. Fiz. Nauk. **167**, 1137 (1997); Sov. Phys. Usp. **40**, 1087 (1997).
- [20] A. Griffith, K. Qu, and N. J. Fisch, Modulation-slippage tradeoff in resonant four-wave upconversion, Phys. Plasmas 28, 052112 (2021).
- [21] K. Estabrook and W. L. Kruer, Theory and simulation of onedimensional Raman backward and forward scattering, Phys. Fluids 26, 1892 (1983).
- [22] C. J. McKinstrie and R. Bingham, Stimulated Raman forward scattering and the relativistic modulational instability of light waves in rarefied plasma, Phys. Fluids B 4, 2626 (1992).