Physics of Plasmas ARTICLE pubs.aip.org/aip/pop

Simulations of non-integer upconversion in resonant six-wave scattering

Cite as: Phys. Plasmas **31**, 053102 (2024); doi: 10.1063/5.0190674
Submitted: 7 December 2023 · Accepted: 13 April 2024 ·
Published Online: 1 May 2024

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ABSTRACT

Resonant upconversion through a sixth order relativistic nonlinearity resulting in a unique resonance was recently proposed [Malkin and Fisch, Phys. Rev. E **108**, 045208 (2023)]. The high order resonance is a unique non-integer multiple of a driving pump frequency resulting in a frequency upshift by a factor of ≈3.73. We demonstrate the presence, unique requirements, and growth of this mode numerically. Through tuning waves to high amplitude, in a mildly underdense plasma, the six-photon process may grow more than other non-resonant but lower order processes. The growth of the high frequency mode remains below the nonlinear growth regime. However, extending current numerical results to more strongly coupled resonances with longer pulse propagation distances suggests a pathway to significant upconversion.

I. INTRODUCTION

The falloff in available laser power below the optical is severe,[1] and producing high intensity UV, XUV, or x-ray pulses remains a challenge with current electron beam driven[2,3] or laser driven sources.[1,4–6] Recent proposals detail using resonant interactions in plasma to upconvert high fluence sources, such as the National Ignition Facility,[7] to shorter wavelength.[8–11] This paper details the first numerical test of higher order plasma mediated wave mixing for upconversion.[12]

Wave mixing builds off of a long history of using plasmas components to replicate the behavior of well-known solid state parts, but which operate at higher laser intensity. Pulse amplification can be done with electron plasma waves,[13–19] ion acoustic waves,[20–22] or magnetized waves.[23] Ion acoustic waves might also enable beam combination.[24–26] Plasmas could serve as polarizers,[27,28] q-plates,[18] lenses,[29] or gratings.[30]

Mildly relativistic wave mixing to upconvert optical light with high efficiency to much shorter wavelengths is a more recent proposal. Pump photons are combined into a higher frequency output through nonlinear coupling. The nonlinear coupling arises from corrections to the plasma frequency at significant electron Lorentz factor.[8] This coupling contrasts with previous work in that it can be resonant, without requiring a modulated plasma density.[31] The lowest order nonlinearity results in four photon coupling. A simple colinear alignment between pump and seed beams requires a complex set of resonance conditions,[9,10] and non-colinear arrangements result in difficulties for amplification.[11] A higher order, six-photon, process addresses both of these problems.[12] This six-photon scattering process consumes four input photons with frequency ω to produce a pair of photons at frequencies of approximately $(2+\sqrt{3})\omega$ and $(2-\sqrt{3})\omega$. This unique non-integer multiple frequency shift from the six-photon process could allow for large changes in wavelength if it were to be cascaded multiple times.

We present a numerical investigation, which demonstrates the unique six-photon resonance. In contrast to previous work, here the resonance is not assumed in a reduced (slowly varying envelope) and expanded model. Instead, we consider a model, equivalent to a cold relativistic electron fluid, in which the full wave dynamics are evolved without the resonance embedded. As the resonance is of higher order, coupling must be maximized to generate observable effects. Thus, we consider the dynamics without expanding in either normalized plasma density, $n_0/n_c = \omega_{pe}^{-2}\omega^2$, for laser frequency ω and plasma frequency $\omega_{pe}^2 = 4\pi e^2 n_0 m_e^{-1}$, or normalized wave amplitude, $e|\mathbf{A}|/mc^2$, for laser vector potential $\mathbf{A}$. When both parameters are not $\ll 1$, which is required to achieve significant coupling, there may be large errors in any expansion of nonlinear terms to finite order. Numerical experimentation is used to examine the fundamental physics at play and validate the viability of relativistic six-wave mixing for upconversion.

The exposition of our numerical results on relativistic six-wave mixing in plasmas is outlined as follows. In Sec. II, we review the dynamics and resonances available for the coupling between mildly relativistic waves, $a < 1$, in an underdense $\omega_{pe} < \omega$ plasma. In Sec. III, we describe the choice of the particular numerical tools used to examine this regime. In Sec. IV, we demonstrate the unique resonance proposed in by Malkin and Fisch. In Sec. V, we discuss the applicability of this work to the aim of achieving significant upconversion, review our work, and discuss possible avenues for continued exploration.

II. BACKGROUND

We review and reformulate how the coupling of electromagnetic waves may be achieved through relativistic corrections to electron dynamics in strong electromagnetic waves. The model in which the coupling occurs is that of an electron fluid, which is driven purely by electromagnetic forces, neglecting electron temperature and pressure along with any ion dynamics. Thus, the fluid is strictly described by Maxwell's equations and the coupled electron quantities of density and velocity. Working in the Coulomb gauge, the dynamics may be described by non-dimensionalized vector potential $\mathbf{a} = e\mathbf{A_x}/m_e c^2$, electrostatic potential $\phi = e\Phi/m_e c^2$, and electron action $s = S/m_e c^2$, resulting in three coupled equations as written by Malkin and Fisch,[9]

$$\left[\partial_t^2 - c^2\partial_z^2 + \frac{\omega_{pe}^2 + c^2\partial_z^2\phi}{\sqrt{1 + a^2 + (c\partial_z s)^2}}\right]\mathbf{a} = 0, \tag{1}$$

$$\frac{\omega_{pe}^2 + c^2\partial_z^2\phi}{\sqrt{1 + a^2 + (c\partial_z s)^2}} c\partial_z s + c\partial_t\partial_z\phi = 0, \tag{2}$$

$$\partial_t s - \phi + \sqrt{1 + a^2 + (c\partial_z s)^2} - 1 = 0. \tag{3}$$

These equations result from combing Maxwell's equations and the relativistic dynamics for electron particle motion, they neglect all kinetic effects under the assumption that the relativistic motion dominates the thermal motion.[9] In the case we shall consider, we assume propagation purely in the z direction, with the vector potential $\mathbf{a}$ only having a transverse, x, component. In this case, the conservation of canonical momentum makes the electron perpendicular momentum $p_\perp = mca$, and the action may be related to the more easily understood quantity of parallel momentum through the relation $p_z = mc^2\partial_z s$. To complete the correspondence between Eqs. (1)–(3) and cold electron hydrodynamics, electron density can be straightforwardly related to the electrostatic potential through Gauss's law, $\omega_{pe}^2 + c^2\partial_z^2\phi = \omega_{pe}^2 n_e/n_0$.

The dynamical equations contain multiple sources of scattering. Raman scattering, where there is coupling between two electromagnetic waves and an electron plasma wave, is the lowest order process. When terms are limited to the second order, fluctuations of density are captured by the electrostatic potential in Eq. (1), which are driven by the ponderomotive force, derived from combining Eqs. (2) and (3) at the lowest order. For the purpose of this paper, Raman scattering is purely parasitic, and forward Raman scattering may grow of off seeded fluctuations in the plasma to move energy into waves, which are no longer resonant for upconversion. The higher order processes responsible for coupling, which may result in upconversion, occur from the expansion of the electron Lorentz factor $\gamma = \sqrt{1 + a^2 + (c\partial_z s)^2}$. The first term in this expansion gives the four-photon coupling,[8] and the next provides the six-photon coupling.[12] These resonances are unique in that they result from the full consideration of the electron dynamics without averaging the ponderomotive force. The subtleties between different resonance regimes and the precise scaling of the coupling in these regimes are not discussed here, as the particular expressions can be extensive and are best understood in a more thorough treatment.

The six-photon scattering process may be characterized by a simple set of resonance conditions and the asymptotic behavior of the coupling. Perpendicularly polarized waves have dispersion relation, written as $\omega_j^2 = k_j^2 + n$, which is normalized for $k_0 = 1$ and $c = 1$. These waves may couple to one another through the relativistic nonlinearity. The fifth order nonlinearity supports several different six-photon scattering processes, but the simplest case consists of one where four input photons at frequency ω_0 and wavenumber k_0 are turned into two photons, one at ω_1, k_1 and one at ω_2, k_2, meeting the following resonance conditions:

$$4\omega_0 = \omega_1 + \omega_2, \quad 4k_0 = k_1 + k_2. \tag{4}$$

These conditions may be solved resulting in the following resonant frequencies:

$$\omega_1 = 2\omega_0 + \sqrt{3}, \quad \omega_2 = 2\omega_0 - \sqrt{3}. \tag{5}$$

The growth of the envelopes of these modes in the system can be represented by a set of dynamical equations in amplitudes, a_j, of

$$-2i(\omega_j\partial_t + ck_j\partial_z)a_j \approx \frac{\omega_{pe}^6}{\omega_0^4} V\partial_{a_j^*}\left(a_0^4 a_1^* a_2^* + a_0^{*4} a_1 a_2\right) \tag{6}$$

for a dimensionless factor V depending on the particularities of the resonance, which is $\sim$1 for the range of parameters considered in this paper. This resonance is unique in that it can occur for co-linear wavevectors in a plasma without renormalization of the laser frequency, which is not possible for lower order relativistic scattering.[9] Furthermore, in the limit, this produces a unique non-integer resonance, in contrast to typical integer harmonics. The growth rate of the six-photon coupling, Γ, normalized to the laser frequency, ω_0, scales as follows:

$$\Gamma \propto (n/n_c)^3 a_0^4 \omega_0. \tag{7}$$

The resulting gain length, L, may be estimated by the expression $L = \frac{V}{\pi}\left(\frac{\omega_0}{\omega_{pe}}\right)^6 \frac{\lambda_0}{a_0^4}$. The form of the leading coefficient and the corrections in higher orders of plasma density may be seen in the exposition by Malkin and Fisch.[12] From Eq. (7), it can be seen that the growth of any six-photon scattering is highly sensitive to both wave amplitude and density.

The parameters to produce a validation of the resonance outline by Eq. (5) are restrictive. To validate the non-integer resonance, without assuming it *a priori* requires a full wave model. This full wave model must have a high resolution to resolve the high frequency mode. However, amplification distances resulting from the weak coupling given by Eq. (7) result in significant plasma and pulse lengths, which are difficult to resolve. To maximize the coupling, we use Eq. (7) to guide the parameter selection toward a high intensity a_0 and high plasma density n. The more highly resolved model allows us to work in a regime where higher order corrections to the dispersion relation and to the equations of motion are more accurately captured than in a nonlinear series expansions or in an envelope based simulation.

III. NUMERICAL METHODOLOGY

The numerical evolution of the relativistic scattering process is chosen to maximize the observability of the six-photon scattering process. To capture a distinguishable signal, the pseudospectral package Dedalus[32] is used in order to minimize numerical noise in the Fourier spectrum, in contrast to high noise particle in cell results. The nonlinear pulse propagation is non-dimensionalized and transformed into the co-propagating frame $\eta = t, \xi = z - t$,

$$\left[\partial_\eta^2 - 2\partial_\eta\partial_\xi + \frac{n_0 + \partial_\xi e}{\sqrt{1 + a^2 + l^2}}\right]a = 0, \tag{8}$$

$$\partial_\eta e - \partial_\xi e + \frac{n_0 + \partial_\xi e}{\sqrt{1 + a^2 + l^2}}l = 0, \tag{9}$$

$$\partial_\eta l - \partial_\xi l - e + \partial_\xi\left(\sqrt{1 + a^2 + l^2}\right) = 0, \tag{10}$$

where $c = 1$, and density is normalized such that $n = \omega_{pe}^2/k_0^2$. Given that only the derivatives of ϕ and s couple to the other equation, for convenience, we reduce the equations with the substitutions $\partial_z\phi = e$ and $c\partial_z s = l$. To ensure numerical stability, a viscosity term is added to curtail the rapid nonphysical growth of high frequency modes driven by the complete absence of physical dissipation.

Simulations are initialized to test for the growth of the proposed six-photon scattering when waves are evolved by Eqs. (8)–(10). In a homogeneous plasma of density of n, we initialize a strong "pump" wave packet with $k_0 = 1$ and a "seed" wave packet of varying k_2. An example of the initial conditions can be seen in Fig. 1. The seed frequency is given a weak amplitude and, to account for different group velocities, is initially leading the pump wave. In the ideal case, the pump wave is completely depleted, and all of the energy is in the low and high frequency waves post interaction. The initial conditions for a, e, and l, are chosen such that the time derivative of each field are initially zero to the lowest non-zero order. Through examination of the spectrum, and in particular, the mode at $k_1 = 4k_0 - k_2$, we demonstrate that Eq. (5) describes a growing mode, which is only seen when it is satisfied.

IV. RESULTS

The presence of a uniquely resonantly growing mode can be demonstrated in the evolution of Eqs. (8)–(10). For a strong laser pulse, many different scattering channels operate coincidently resulting

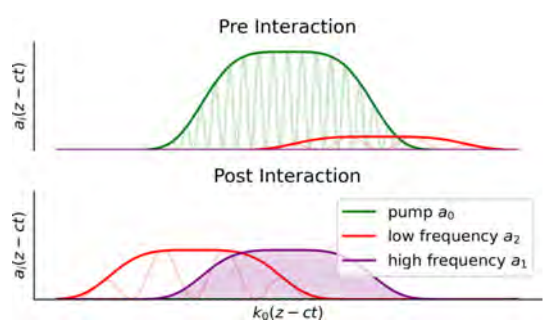

FIG. 1. The pump wave is initialized with a super-Gaussian envelope, which is led by a much weaker low frequency seed. If pump depletion could be reached, the ideal outcome would be such that the high frequency wave and low frequency wave are amplified until the pump is completely depleted. While at equivalent amplitude, the high frequency wave would contain the majority of the remaining energy due to having a much higher frequency.

in the growth of a large number of discrete modes. The intensity of these modes is oscillatory, unless conditions are precisely chosen such that waves are pushed into resonance.

The nature of this resonance can be demonstrated by varying the wavenumber, k_2, of a low frequency seed and examining the energy that is scattered into the expected resonance at $4k_0 - k_2$. Both waves start in a homogenous plasma and are evolved until a minority of the seed overlaps with the pump, as diagramed in the co-propagating coordinate scheme in Fig. 1. The plasma is initialized such that $k_0/k_{pe} = 7.5$. While the plasma density is such that it is relatively underdense for the pump wave, we note that the large frequency separation between the pump and the seed wave means the plasma density is not much smaller than the quarter critical density of the seed wave. The pump is given an $a_0 = 0.5$, and the seed is overlapped with a factor of 200 lower intensity. The pulse length, which is set by the slippage condition between the pump and the seed is 450 wavelengths, which propagates for a duration of 4000 laser periods. This propagation length is much shorter than the estimated gain length of 8×10^5 laser wavelengths, and reaching the nonlinear regime will not be possible with the current code. This duration is set such that at the end of the simulation, the low frequency seed will have slipped past half of the pump wave, at which point the growth of the high frequency mode has been arrested. The seed frequency is varied around the proposed resonance, with a minimum frequency bounded by two plasmon decay $\omega_2 = 2\omega_{pe}$, and the maximum frequency bounded by the coupling of the third harmonic of the low frequency seed to the stokes shifted pump wave, where $3\omega_2 = \omega_0 - \omega_{pe}$. Both processes at the bounds are lower order and quickly produce a large electron plasma wave, whose dynamics dominate the desired higher order upshift.

The evolution of the simulation spectrum shows the many nonlinear processes at play, but the resonant six-photon process is the most significant. The spectrum created by the wave simulation for a resonant simulation is shown in Fig. 2. For a resonant k_2, the strongest growth is of the six-photon resonance; however, energy may be transferred to many other modes. For example, the second most defined new peak in the spectrum is the third harmonic of the pump wave. However, even though this mode is created by a lower order and thus more strongly coupled, it is not resonant so it cannot continually grow and is saturated at this level. The evolution of the energy in different linear combinations of the pump, seed, and plasma frequency shown in Fig. 2 demonstrates this. Other bands oscillate in energy, while the high frequency six-photon resonance grows to a level higher than that of other nonlinear processes. The growth of the six-photon resonance shows good agreement with the analytical theory, where the analytical estimate is $1.6 \times 10^{-7}\omega_0$ and a fit to the growth in simulations gives a growth rate of $1.8 \times 10^{-7}\omega_0$. The growth rate peaks when there is good overlap between the pump and the seed, declining at later times as the seed spatially lags behind the pump wave. Many other peaks corresponding to combinations of the pump and seed wave occur in the spectrum, but at weaker levels. Of particular note is the four-photon process, corresponding to $k = 2k_0 - k_2$, which experiences some scattering, but is not resonant in the colinear geometry without further tuning of the pump and seed wave.[9]

Varying the seed frequency demonstrates the uniqueness of the resonance point. Figure 3 shows the energy in the post interaction spectrum for simulations at varying seed frequency, k_2. Only the nonlinear growing modes are presented as the pump and seed wave are

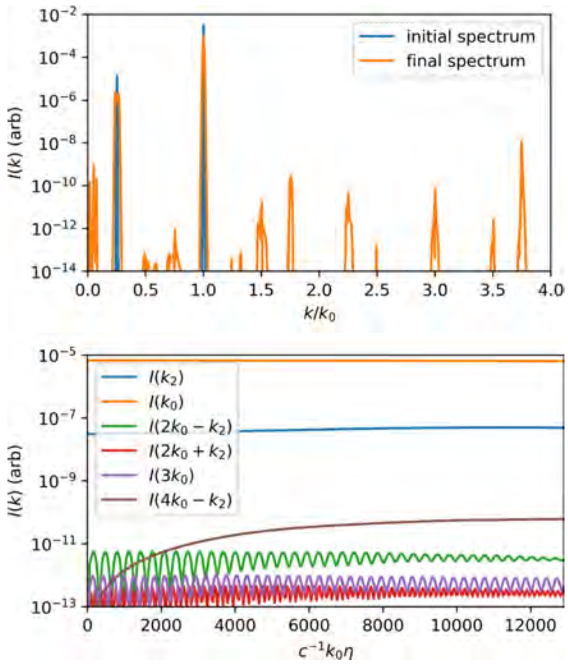

FIG. 2. The evolution of the spectrum of a pump ($k_0 = 1$, $a_0 = 0.5$) and seed [$k_2 \approx (2 - \sqrt{3})k_0$, $I_2 = I_0/200$]. Both waves are initialized within a homogenous plasma and evolved until a majority of the seed has slipped past the pump. Nonlinear distortions of the pump and seed are weak, but many peaks can be seen in the final spectrum compared to the initial spectrum (top). In k space, many spontaneous, but non-growing, modes are instantaneously excited across the spectrum. The particular mode of interest at $4k_0 - k_2$ may grow if Eq. (5) is satisfied allowing it to achieve higher intensities than those at modes resulting from lower order processes. This can be seen through looking at the energy in each frequency band, (bottom), where only the resonant mode is growing, and other nonlinearly driven bands oscillate in energy. The energy for each band is calculated by integrating the spectrum in a window of each peak of width $k_{pe}/4$.

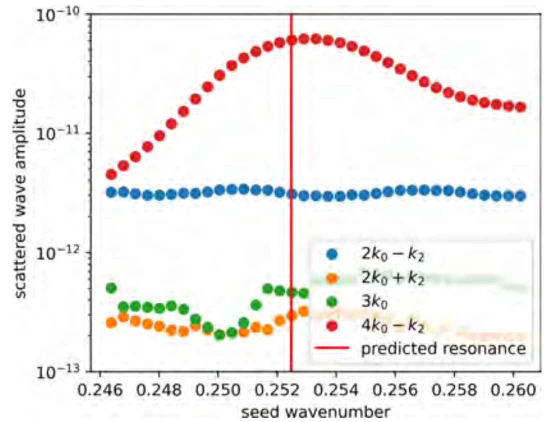

FIG. 3. A parameter scan across seed wavenumber k_2 results in the varied excitation of many modes. Each scatter point corresponds to the energy in the final spectrum for the dominant nonlinear modes. The expected resonance at $k_2 = 4k_0 - [(2\sqrt{1+n} + \sqrt{3})^2 - n]^{1/2} k_0 = 0.252k_0$ is more highly amplified than neighboring wavenumbers. The range of the parameter scan over k_2 is heavily restricted by the growth of electron plasma waves at the bounds of k_2. Shorter duration simulations highlight a larger difference in the energy of the amplified across a wider range of k_2 at the cost of less amplification of the k_1 wave.

strong enough to dominate the weak nonlinear modes. The strongest nonlinearly created mode is the predicted $4k_0 - k_2$ mode. The parameter scan is restricted by other resonances in the system. As the seed frequency approaches the upper value plotted, the third harmonic of the seed approaches the Stokes line of the pump, resulting in significant Raman scattering.

Some initial small amount of energy can be transferred into the $4k_0 - k_2$ mode regardless of the seeded k_2. However, this energy will oscillate if k_2 is not chosen correctly, which occurs as the seed frequency is moved away from the resonance, shown by the red line. As k_2 becomes closer to the resonance, the period of oscillation grows, and the energy may coherently grow. The efficiency of this process remains low, with the energy in the high frequency band more than 10^{-5} below the initial pump energy. To reach the nonlinear growth regime, the resonance has the condition $\delta\omega_2/\omega_2 \lesssim \frac{1}{2}\frac{n^2}{\omega_0^4} a_0^4$. For the parameters in the simulations presented, this corresponds to a maximal detuning of 10^{-5}. This is stricter than the results from Fig. 3 as the simulations are not propagated for a long duration. Achieving more significant growth, and correspondingly lowering the requirements on the resonance, will require a significantly greater plasma density.

Other resonances may result in growing parasitic modes and become limiting. The desired scattering becomes dominated by Raman scattering if the seed frequency can create an electron plasma wave. Raman scattering may be seeded off of noise; however, there are two other limits also present as the seed frequency changes. If the seed frequency approaches the threshold for two plasmon decay at low k_2, or the third harmonic of the seed approaches the Stokes shifted frequency of the pump, Raman scattering may become strong. At these modes, the spectra starts to produce many ω_{pe} shifted bands, which distort the spectrum. When these two resonances occur, the electron plasma wave, and corresponding density fluctuations, quickly grow beyond the limits of the cold hydrodynamic model. We aim to avoid Raman scattering, so this limitation should not be relevant for k_2 detuned from these operating points. However, at high density, these bounds may be close to the resonance where $\omega = 4\omega_0 - \sqrt{3}k_0$. This limits the plasma density, and thus the coupling of the six-photon coupling. Fluctuations in plasma density may push the low frequency wave into resonance and produce a large electron plasma wave. A noisier environment than the shown simulations will also provide a more fecund environment for parasitic Raman scattering. Further manipulation of the plasma and laser pulses will be needed such that Raman scattering does not dominate any desired higher order nonlinearity.

V. SUMMARY AND DISCUSSION

This paper demonstrates through numerical simulation the unique non-integer harmonic resonance driven by six-photon coupling in a plasma. This coupling, when at resonance, driven by the fifth order corrections from the electron Lorentz factor, is large enough that in the post interaction spectrum, the mode is larger than other nonlinear processes. This is true even though Raman scattering and relativistic four-wave mixing are lower order processes. It is isolated in low noise pseudospectral simulations. However, for the pulse durations and propagation distances considered, it is still below the strength of the injected waves. The non-integer multiple of the frequency is required for the six-photon scattered wave to be strong. If the frequency is detuned, growth of the high frequency mode is orders of magnitude lower, and if it is significantly detuned, the injected wave can drive other parasitic processes.

The six-photon process is distinguishable at demonstrated parameters, but reaching efficient upconversion will require further modeling. The pulses in this paper have a picosecond duration and propagate through 2 mm of plasma, producing a high frequency wave of 10^{-5} lower intensity than the pump. This is highly inefficient, but, given the significant pump energy, might be detectable in a sensitive experiment. There are challenges that must be addressed before this can be accomplished. While we have considered how waves in the system might seed the growth of plasma waves, our treatment has assumed that electrons are perfectly cold. Kinetic effects may significantly seed the growth of parasitic plasma waves, and it may be difficult to find a parameter regime in which these effects are balanced against significant growth. More detailed study will need to be done in higher growth regimes to examine the balance of these effects. Growth falls short of the requirements for application, where scaling the growth length results in a plasma channel length that is unachievable. This is primarily because a lower plasma density has been used than in the most promising upconversion schemes.[12] The use of high densities and high growth has been limited by the development of plasma instabilities, which might be overcome in further simulation. Inhomogeneous plasma conditions, which disrupt the growth of plasma waves, could allow density, and thus coupling to be much higher. Even in a higher coupling regime, pulses will propagate for many more laser cycles. Resolving the shortest wavelengths and highest frequencies would be costly, but can be overcome by working with an envelope model, which would only need to resolve envelope length scales and nonlinear process timescales.

Working around these challenges to model and, eventually, experimentally produce a high energy, short wavelength pulse could be done through the application of a more sophisticated approach. Notably, the six-photon resonance here, which uses a single pump, has a much lower growth rate than those consisting of detuned pump waves. When the beating of the pumps is chosen precisely, the resonance achieves a smaller frequency multiplication, but with much higher growth.[12] Parasitic Raman scattering might also be suppressed through varying the density of the plasma. Varying the density would be favorable, as the six-photon resonance considered here results from the coupling of electromagnetic waves, which are less sensitive to the plasma density than Langmuir waves; thus, the resonance is less sensitive than the Raman resonance. Though it is important to note that this will not remain the case if the beat wave between pumps becomes relevant for other six-photon processes. Higher order process requires a high intensity over a large propagation distance, so filamentation may become a problem if low diffraction and, thus, high power is needed. If these conditions are met, simulations performed using particle-in-cell methods could directly show the six-photon process taking into account kinetic effects. Six-photon scattering contains great promise, and iterated scattering, which might be automatically seeded, could produce geometric increases in frequency.

ACKNOWLEDGMENTS

The authors thank Vladimir Malkin for informative discussions. This research was supported by the NSF PHY-2206691, DOE DE-SC0021248, and DOE NNSA DE-NA0004167.

AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Alec Griffith: Investigation (equal); Methodology (equal); Software (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Kenan Qu:** Investigation (equal); Supervision (equal); Writing – original draft (equal); Writing – review & editing (equal). **Nathaniel J. Fisch:** Conceptualization (equal); Funding acquisition (equal); Project administration (equal); Supervision (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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