Stimulated-Raman-scattering amplification of attosecond XUV pulses and application to local in-depth plasma-density measurement

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(Dated: July 15, 2022)

We present a scheme for amplifying an extreme-ultraviolet (XUV) seed isolated attosecond pulse via stimulated Raman scattering of a pulse-train pump. At sufficient seed and pump intensity, the amplification is non-linear and the amplitude of the seed pulse can reach that of the pump, one order of magnitude higher than the initial seed amplitude. In the linear amplification regime, we find that the spectral signature of the pump pulse train is imprinted on the spectrum of the amplified seed pulse. Since the spectral signature is imprinted with its frequency downshifted by the plasma frequency, it is possible to deduce the electron density in the region of interaction, which can be as short as micrometer longitudinally. By varying the delay between the seed and the pump, this novel scheme provides a local electron-density measurements inside solid-density plasmas that cannot be probed with optical frequencies, with micrometer resolution.

I. INTRODUCTION

With high-order harmonic generation in gas target (gas-HHG) [1, 2], it is possible to generate isolated attosecond pulses (IAP) in the extreme-ultraviolet (XUV) regime [3–6]. Such pulses can be used as probes to study processes on the attosecond timescale [7], e.g. electron wave-function dynamics in atoms and molecules [8–10], as well as plasma diagnostics [11–15]. However, these pulses are relatively low energy and will, in general, not be able to significantly drive a plasma in an XUV-pump–XUV-probe experiment [16, 17].

Another XUV source is high-order harmonic generation in laser–solid interactions [18] (solid-HHG), either via surface plasma waves [19, 20], coherent synchrotron emission [21–23], the relativistic electronic spring [24, 25], or the relativistically oscillating mirror mechanism [26–31]. The latter mechanism has the potential to provide XUV pulses with very high intensities, potentially up to \( I \sim 10^{25} \text{Wcm}^{-2} \) [29]. With these methods, a train of high-intensity attosecond XUV pulses can be generated with sufficiently high field strength to significantly affect the electrons and drive collective processes in a plasma. There are also methods for generating IAPs with solid-HHG [22, 32–37].

In this paper we propose the use of stimulated Raman back-scattering (SRS) [38–43] to amplify an IAP seed pulse with a pulse-train pump. While the three-wave-coupling system of equations for SRS is fairly well understood for pseudo-monochromatic pulses, its applicability to the broad-spectra IAP seeds and pulse-train pumps considered here is not obvious. We therefore explore the possibility of this approach via particle-in-cell (PIC) simulations, and present parametric studies of the SRS amplification with respect to initial pump and seed amplitudes as well as pulse-train length.

Since SRS relies on the presence of electron-plasma waves induced by the pump and seed laser pulses, the efficacy of the amplification is highly dependent on both the pump and seed normalized amplitudes \( a_0 \) and \( a_1 \), respectively. Our simulations indicate that efficient non-linear amplification, without severe temporal stretching of the seed pulse, requires \( a_1 \gtrsim 0.02 \), which is almost within reach for gas-HHG, with reported amplitudes of \( a_1 \sim 0.01 \) [6].

An interesting application of this XUV–SRS scheme is for in-depth, local electron-density diagnostics. Because high-density plasmas are opaque to optical frequencies, light-based probing methods have to be in the XUV or x-ray range of wavelengths. Previous experiments have utilized XUV transmission [11, 12] to determine electron densities on ~100fs timescales. Furthermore XUV wavefront sensing [13] and dispersion of XUV pulses [15] have been proposed to diagnose solid-density laser-generated plasmas on the timescale of the XUV pulse. However, these techniques all produce line-integrated measurements along the path of the probe pulse. By instead utilizing the locality of the SRS interaction, the method proposed in this paper has the potential to probe the plasma density, inside the plasma, with micrometer-scale longitudinal resolution by varying the delay between the pump and seed.

The proposed local electron-density diagnostics method is based on the spectral imprinting of the spectral fringes of the pump pulse train onto the broad-spectrum seed pulse. Since a pulse train has a spectrum consisting of several prominent peaks (depending on the separation of the pulses in the train) and the SRS coupling is strongest at a frequency shift equal to the plasma frequency \( \Delta \omega = \omega_p \), these spectral peaks will be
imprinted onto the spectrum of the seed pulse. Thus, the plasma frequency, \( \omega_p \), and thereby the electron density, can be deduced from measuring and comparing the spectra of the pump and amplified pulses. Note that this spectral imprinting is most efficient in the linear SRS amplification regime, i.e. for \( a_1 \lesssim 0.01 \), which is currently realizable.

II. INTERACTION REGIMES

Consider a pump and a counterpropagating seed in a plasma with frequencies \( \omega_0 \) and \( \omega_1 \), respectively, fulfilling the phase matching condition \( \omega_0 - \omega_1 = \omega_p \), where \( \omega_p \) is the plasma frequency; the pulses have normalized amplitudes \( a_i = e E_i / (m_e c \omega_i) \) (for \( i = 0, 1 \)) with \( E_i = \sqrt{2 I_i / \epsilon_0} \) and where \( e \) is the elementary charge, \( m_e \) the electron mass, \( c \) the speed of light, and \( \epsilon_0 \) is the permittivity of free space, and \( I_i \) is the intensity of the respective beams. Following Edwards et al. [44], the linear-regime SRS growth rate \( \Gamma \) for the seed pulse inside a plasma with electron density \( n_e \) is given by

\[
\frac{\Gamma}{\omega_0} = \frac{1}{2} \left[ \left( \nu_3 - \nu_1 \right)^2 \nu_3 + \nu_1 \right]/2, \tag{1}
\]

where

\[
\hat{\nu} = \frac{a_0 e \left( |k_0| + |k_1| \right) (n_e/n_{e,0})^{1/4}}{4 \sqrt{2} \omega_1} \tag{2}
\]

is the undamped SRS growth rate with pump and seed wavenumbers \( k_0 \) and \( k_1 \), respectively, and \( n_{e,0} \) is the critical density associated with \( \omega_0 \); \( \nu_3 = \nu_{3,ld} + \nu_{ei}/4 \) and \( \nu_1 = (n_e/n_{e,0}) \nu_{ei} \omega_0 / \omega_1 \) using the normalized collisional and Landau damping rates

\[
\nu_{ei} = \frac{2 \sqrt{2}}{3 \sqrt{\pi n_e / n_{e,0}}} \frac{e^2 \omega_0 \log A}{4 \pi e^2 m_e c^3 q_e^{3/2}} \tag{3}
\]

and

\[
\nu_{3,ld} = \frac{\sqrt{\pi n_e / n_{e,0}}}{(2 q_e)^{3/2}} \exp\left(-1/q_e\right), \tag{4}
\]

respectively, where \( q_e = 4 T_e / (m_e c^2 n_e / n_{e,0}) \), and the Coulomb logarithm is taken as \( \log A = \log(12 \pi \lambda_D^2 n_e / Z_i) \) with the electron temperature \( T_e \), Debye length \( \lambda_D \) and ion charge number \( Z_i \).

In this paper we study pump amplitudes ranging from \( a_0 = 0.05 \) to 0.2, which results in growth rates in the order of \( \Gamma / \omega_0 \sim 0.023 - 0.1 \) at \( n_e \approx 0.6 n_{e,0} \). As discussed in Edwards et al. [44] for wavelengths above 10 nm and for the pump amplitudes considered here amplification by SRS should be more favorable than SBS, or in some case the difference will be marginal. However, this theory assumes quasi-monochromatic pulses, which is not the case in our study. We have also verified, by complementary simulations (not shown here), that indeed the SBS amplification is not efficient for our setup and parameter range. As it is expected that the observed growth rates should be lower due to the “gaps” between pulses in the pump pulse train, the faster time scales associated to SRS are essential. Furthermore, due to the short pulse durations, the energy in the pump and seed pulses are spread over a broad spectral range, which is expected to differently affect the growth rates of the spectral components of the seed. To our knowledge, there is no developed fully spectral theory of the SRS with broad-band pulses, but we expect that the growth rate of each spectral component is approximately proportional to the spectral density \( |\hat{E}| \) of the pump – similar to the \( a_0 \) dependence in (2) – where \( \hat{E} \) is the Fourier transform of the electric field.

It has been pointed out that in order to have efficient SRS amplification, it is useful to start with a seed that has initial amplitude and duration close to the optimal condition of non-linear amplification [43], namely \( a_1 T \omega_0 (n_e/n_{e,0})^{1/2} \approx 6.8 \), where \( a_1 \) and \( T \) are respectively the amplitude and duration of the seed pulse once the non-linear regime has set in, with \( a_1 \) comparable to \( a_0 \). Typical seed pulses in the XUV range will be only a few cycles, and will interact in relatively high densities plasmas, of the order of 0.1 – 0.2 \( n_{e,0} \). In order to fulfill the condition above, we would need a very large initial seed amplitude \( a_1 = 0.5 \), that is beyond the parameter range of interest for this paper. Nonetheless, for sufficiently large pump intensity \( (a_0 \gtrsim 0.2) \) we do enter in a nonlinear regime of amplification as will be discussed below.

III. SIMULATION SETUP

We used the Smilei PIC code [45] to simulate the interaction between a seed pulse and a pump pulse train in a plasma in one dimension (1D). The simulations were performed in various box sizes, all with a spatial resolution of \( \Delta x \approx 0.7 \) nm, at least 50 times smaller than the laser wavelengths used in this study. The seed pulse considered here had a central wavelength of \( \lambda_1 = 50 \) nm and the Gaussian temporal envelope with a (field-amplitude) full-width-at-half-maximum (FWHM) duration of four cycles. The plasma was chosen to be a pure hydrogen plasma at densities \( 0.1 n_{e,1} \) (Sec. IV A) and \( 0.2 n_{e,1} \) (Sec. IV B), where \( n_{e,1} \) is the critical density associated with the seed central frequency \( \omega_1 \). The plasma was modeled using 1000 and 800 particles per cell for the electrons and protons, respectively. The initial electron and ion temperatures were \( T_e = 100 \) eV and \( T_i = 1 \) eV, respectively. The pulse trains considered had central wavelengths of \( \lambda_0 = 38 \) nm (Sec. IV A) and \( \lambda_0 = 36 \) nm (Sec. IV B); the individual pulses in the train were also four-cycles FWHM duration – based on their own central wavelength. The distance between pulses in the train was 400 nm.

We note that the use of 1D PIC simulations is well justified under these conditions. The fastest-growing
transverse instability one can expect, the filamentation instability [46, 47], grows over hundreds of femtoseconds time scales and sub-micrometer transverse scales, much larger than those considered in this work.

IV. RESULTS AND DISCUSSION

We first discuss the amplification of an IAP (Sec. IV A), considering in particular the effect of the lengths of pump pulse trains. Most efficient amplification, without severely extending the duration of the amplified seed pulse, is shown to occur in the non-linear regime. In contrast, in the linear SRS regime (Sec. IV B), the clear spectral signature of the pump pulse train is imprinted onto the seed pulse with a frequency shift equal to the local plasma frequency. By comparing the spectra of the amplified and pump pulses, this technique can be used to deduce the electron density inside the plasma, with micrometer resolution.

A. XUV-pulse amplification

Figure 1 shows the amplification of a seed pulse with initial amplitude \( a_1 = 0.02 \); panel (a) shows the pulse before and panel (b) after interaction with the pump pulse train consisting of \( N = 10 \) pulses with wavelength \( \lambda_0 = 38 \text{ nm} \). The black dotted curves in panel (a) and (b) indicate the spatial filtering used to obtain the spectra of the seed pulse shown in Fig. 1(c), where the dashed blue line represent the spectrum of the initial seed pulse and the solid red line represent that of the amplified seed pulse; the dotted magenta line show the spectrum of the pump after SRS has occurred. The vertical dashed lines show the spectral maxima of the pump and amplified seed pulses; the corresponding frequency downshift \( \Delta \omega \) agrees very well with the plasma frequency \( \omega_p \) associated with the electron density \( n_e = 0.1 n_{c,1} \) used here.

By integrating the spatially filtered spectra, shown in Fig. 1(c), we can calculate the energy \( \mathcal{E}_1 \) of the seed pulse before and after amplification, which allows us to compute the relative energy gain \( \mathcal{G} = \mathcal{E}_1^\text{final}/\mathcal{E}_1^\text{init} \simeq 245 \) in this case. Including collisions in these simulations reduces the energy gain by only \( \lesssim 10 \% \). However, in foil targets the higher ion charge at solid density may result in more significant collisional effects [48].

We have also performed similar calculations for various pulse-train lengths \( N \) with different combinations of pump and seed amplitudes, \( a_0 \) and \( a_1 \), respectively, which are shown in Fig. 2. In the cases displayed, the gain rises rapidly with the number of pump pulses for small \( N \). However, at a certain point the gain saturates at approximately \( \mathcal{G} \sim (a_0/a_1)^2 \), which roughly corresponds to the seed pulse reaching the amplitude of the pump. Extending the pump pulse train beyond this saturation point mostly results in a broadening of the pulse envelope and an increased background.

![Figure 1](image1.png)

**FIG. 1.** Amplification of an IAP by an XUV pulse train via non-linear SRS, with initial pump and seed amplitudes \( a_0 = 0.2 \) and \( a_1 = 0.02 \), respectively. Real-space field (relativistically normalized with the central frequency of the seed) before (a) and after (b) the seed–pump interaction. The dotted black lines indicate the spatial filtering envelopes used to distinguish the spectra of the seed/amplified pulse. (c) Spectra of the amplified (solid red line) and initial seed pulse (dashed blue line), as well as the pump pulse train after (dotted magenta line) interaction with the seed. The vertical dashed lines indicate the spectral maxima of the pump and amplified seed pulse.

![Figure 2](image2.png)

**FIG. 2.** Relative energy gain \( \mathcal{G} \) as a function of number of pulses \( N \) in the pump train. The seed and pump pulses are both four-cycle FWHM duration with seed and pump wavelengths \( \lambda_1 = 50 \text{ nm} \) and \( \lambda_0 = 38 \text{ nm} \), respectively. The plasma density is \( n_e = 0.1 n_{c,1} \).
of spontaneous Raman scattering (RS). The unwanted spontaneous RS is especially prominent in the case with $a_0 = 0.2$ and $a_1 = 0.02$ – due to the stronger non-linear effects at higher pump amplitudes – for pulse trains longer than $N > 10$. Already at $N = 10$, there is noticeable spontaneous RS, as seen by the noise floor behind the pulse train in Fig. 1(b).

The stronger non-linearity present at higher pump and seed amplitudes is, however, also key to producing short amplified pulses. As a comparison, the two series with $a_0 = 0.1$ from Fig. 2 generate two to three times longer duration output pulses than the $a_0 = 0.2$ case for $N \leq 10$. To give concrete examples, the amplified-pulse duration with $(a_0, a_1) = (0.1, 0.02)$ and $N = 10$ is $\tau_{95\%} \simeq 6.3$ fs, which can be compared with $\tau_{95\%} \approx 2.7$ fs in the corresponding case with $(a_0, a_1) = (0.2, 0.02)$ [Fig. 1] – the initial seed duration was $\tau_{95\%}^{\text{init.}} \approx 0.8$ fs in both cases – where $\tau_{95\%}$ is measured as the time span that contains 95% energy of the waveform. Furthermore, the $a_0 = 0.1$ cases have lower gain than the $a_0 = 0.2$ case, which indicates that the pump amplitude has a lower threshold for efficient non-linear amplification at $a_0 \gtrsim 0.2$.

We further investigate the saturation of the relative energy gain by varying the initial electron temperature, which affects the strength of Landau damping, as well as using a seed pulse with $a_1 = a_0 = 0.2$, to study whether electron-wave breaking may be causing the saturation. When the electron temperature is varied between $T_e = 50$ eV and 500 eV, the spontaneous RS is somewhat damped. However, the desired stimulated RS is more strongly reduced at higher temperatures. Interestingly, however, including collisions (investigated at temperatures $T_e = 50$ eV, 100 eV and 300 eV) noticeably reduces spontaneous RS, more strongly than the reduction of the desired stimulated scattering, indicating that collisional damping has a larger effect than Landau damping on the spontaneous RS.

Next, in a series of simulations with $a_1 = a_0 = 0.2$, varying the number of pulses in the pump train from $N = 2$ to 10, to study the effect of electron-wave breaking. The result of the $N = 10$ case is shown in Fig. 3, where we in panel b see that the amplitude of the amplified pulse is almost 2.5 times higher than the initial seed amplitude, without any significant increase in pulse duration – the amplified-pulse duration is $\tau_{95\%} \approx 1.0$ fs, compared to the initial seed duration of $\tau_{95\%}^{\text{init.}} \approx 0.8$ fs. While the electron phase space (not shown) in the $a_1 = a_0 = 0.2$ cases show clear tendencies of electron-wave breaking – which occurs when the amplitude of the electron wave is sufficiently large that the fastest electrons significantly outspeeds the wave itself – we still find that the absolute energy gain $\Delta E_1 = E_{1}^{\text{final}} - E_{1}^{\text{init}}$ is approximately twice that of the $(a_0, a_1) = (0.2, 0.02)$ case. Thus, since there is no deterioration of the energy transfer for $a_1 = a_0 = 0.2$, we may conclude that electron-wave breaking is not significantly affecting the non-linear amplification in the cases considered here.

![Figure 3](image_url)

FIG. 3. Amplification of an IAP by an XUV pulse train with the same amplitude seed and pump $a_0 = a_1 = 0.2$. Real-space field (relativistically normalized with the central frequency of the seed) before (a) and after (b) the seed-pump interaction. The dotted black lines indicate the spatial filtering envelopes used to distinguish the spectra of the seed/amplified pulse. (c) Spectra of the amplified (solid red line) and initial seed pulse (dashed blue line), as well as the pump pulse train after (dotted magenta line) interaction with the seed.

### B. Linear SRS as a local density diagnostics

Although the relative energy gain is lower and the amplified pulse duration is longer in the linear compared to the non-linear regime, the same mechanism that causes these undesirable amplification properties can instead be exploited for measuring the electron density inside a plasma. Figure 4 shows the results of the interaction between an $a_0 = 0.1$ pump pulse train and an $a_1 = 0.01$ seed pulse with central wavelengths (in vacuum) of $\lambda_0 = 36$ nm and $\lambda_1 = 50$ nm, respectively. Panels (a) and (b) show the pulses before and after interaction, respectively. We find that the amplitude of the seed had not been greatly affected by the SRS, however, the pulse duration has increased to $\tau_{95\%} \approx 4.6$ fs – practically to that of the full pump pulse train.

The cause for this result is elucidated by the spectra shown in Fig. 4(c). The amplified pulse has acquired three distinct peaks (solid red line) on top of the initial spectrum (dashed blue line). These peaks correspond to a downshift by $\omega_0$ of the three highest peaks of the pump spectrum (dotted magenta line). Spectrally, the amplified seed pulse has become more similar to a pulse train, which is also seen in the real-space shape of the envelope [Fig. 4(b)]. Owing to the frequency shift, the spectral peaks of the amplified seed pulse are still
amplification also depends on the spectral density of
the seed, $|\hat{E}_y^{(1)}(\omega)|$, the seed pulse spectrum should be
sufficiently broad to capture the imprinting of an as wide
as possible section of the pump spectrum, even when the
central frequencies are not perfectly matched – as is the
case in the simulation shown in Fig. 4. If the seed and
pump had narrow spectra, then the frequency difference
between the seed and the pump would have to be swept,
in order to find the largest amplification, before the value
of $\omega_p$ could be established, which could be challenging
experimentally. Of course, the method is still limited in
what plasma densities can be measured – based on
which seed and pump central frequencies are available –
even with a broad-band seed and pump, but the range of
densities that can be probed increases with the seed and
pump bandwidths.

Next, because the spectral imprinting only occurs
while the seed and the pump interact, the probed
frequency shift corresponds to the plasma frequency
only in the interaction region, the size of which is
approximately half the length of the pump pulse train.
While the linear regime operates with relatively low-
amplitude pulses, it should be noted that if the pump
amplitude is below $a_0 \lesssim 0.05$, SRS growth rate becomes
low – even if the seed amplitude is increased to $a_1 = a_0$ –
and a longer pump pulse train is needed, thus increasing
the interaction region. By shifting the pump–seed delay,
it is possible to choose where in the plasma to probe
the density. This method, therefore, has the potential to
probe the electron density at a micrometer longitudinal
resolution, at depth inside the plasma. Depending on the
transverse size of the focus, the probed volume can be as
small as a few tens of \(\mu m^3\).

V. CONCLUSIONS

We have presented a scheme for amplifying isolated
attosecond XUV pulses based on stimulated Raman
scattering. In order to efficiently amplify the attosecond
seed pulse without severely extending its duration, the
seed and pump must be sufficiently strong to enter
the non-linear regime. In particular, we find that the
relativistic amplitude of the seed should be $a_1 \gtrsim 0.02$
with a pump of amplitude $a_0 = 0.2$.

In the linear RS regime, the broad spectrum of the
attosecond seed is imprinted with the peaked spectrum
of the pump pulse train. Because the imprinting of
the pump spectrum occurs with a frequency downshift
equal to the local plasma frequency $\omega_p \propto n_e^{1/2}$, the local
plasma density can be deduced by comparing the spectra
of the pump and amplified seed pulses. The advantage
of this novel, in-depth density-measurement method is that,
depending on the length of the pump pulse train, the
longitudinal spatial resolution can be in the micrometer
scale, allowing for local density diagnostics of solid-
density plasmas inside the plasma.

FIG. 4. Illustration of the spectral imprinting from linear SRS, with initial pump and seed amplitudes $a_0 = 0.2$ and
$a_1 = 0.02$, respectively. Real-space field (relativistically normalized with the central frequency of the seed) before (a)
and after (b) the seed–pump interaction. The dotted black lines indicate the spatial filtering envelopes used to distinguish
the spectra of the seed pulse. (c) Spectra of the amplified (solid red line) and initial seed pulse (dashed blue line), as
well as the pump after (dotted magenta line) interaction with the seed. The vertical dashed lines indicate the spectral
maxima of the pump and amplified seed pulse, separated by a frequency shift of $\Delta \omega \approx 16.8$ fs$^{-1}$.
ACKNOWLEDGMENTS

The authors are grateful for fruitful discussions with P. Eng-Johnsson, A. Gonoskov, T. Fülöp. This project has received funding from the Knut and Alice Wallenberg Foundation (Dnr. KAW 2020.0111). A.S. gratefully acknowledges the support from Adlerbertska forskningsstiftelsen. The computations were enabled by resources provided by the Swedish National Infrastructure for Computing (SNIC), partially funded by the Swedish Research Council through grant agreement no. 2018-05973.

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