Collective effects in the radiative decay of the $^1P$ state in helium

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We present a theoretical study of He $^1P$→$^1S$ emission from a helium gas, where the upper atomic state is prepared by a 100 fs laser pulse resonant with the $1^S$→$1^P$ transition. The fluorescence signal emitted in the forward direction at 2059 nm and the transmitted signal at 58.4 nm are followed in time starting with the low pump intensity regime where the single-atom spontaneous emission results in a characteristic exponential decay. By increasing the gas density and pump intensity, collective effects such as self-amplified spontaneous emission and superfluorescence modify the temporal dependence of the radiation field by amplifying the radiation pattern as well as changing its delay and intensity distribution. The extended Maxwell-Bloch simulation of the corresponding three-level $\Lambda$ system in one spatial dimension covers emission delays of up to 2.4 ns, reaching beyond the natural lifetime of the $^1P$ state.

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I. INTRODUCTION

At present, only free-electron laser (FEL) facilities can provide spectroscopically sharp, ultrashort, intense, and coherent radiation pulses in the extreme ultraviolet spectral region [1]. A passage of such a pulse through a gaseous medium may strongly modify its temporal profile and lead to the specific dependence of the emission field typical of self-amplified spontaneous emission (SASE) or superfluorescence (SF) [2]. Due to the collective response of the atomic ensemble to the radiation field, the fluorescence signal strongly deviates from the well-known exponential time dependence of the single-atom case.

Recently, SF has been observed in a helium gas at 502 nm, corresponding to the $^3P$→$^1S$ transition [3]. At the Spring-8 Compact SASE Source (SCSS), the gas was excited by 100 fs pump pulses in resonance with the $1^S$→$3^P$ atomic transition. Typical SF trends were reported in the 300–3000 Pa pressure range for pump intensities reaching $3\times10^{12}$ W cm$^{-2}$; higher density of excited-state atoms leads to proportionally narrower pulses, higher peak intensities, and shorter emission delay times. To explain the data set obtained by spectrally broad and partially coherent SASE pulses, elaborated simulations of emission cascades were performed that went beyond the simplest two-level case, but left the pump and the decay stage of the process uncoupled [4–6].

On the other hand, SASE at shorter wavelengths (1.46 nm) was first demonstrated at the LCLS FEL facility using 40–80 fs x-ray pulses focused on the gas cell filled with $7\times10^4$ Pa of neon [7]. The central wavelength of the broadband SASE pulses was tuned to 960 eV to generate an inverted population of $K$-hole ions. With the pump intensity approaching $2\times10^{12}$ W cm$^{-2}$, an exponentially increasing $K_a$ emission yield at 849 eV was observed in the forward direction. Recently, SASE was observed also at shorter wavelengths, examining the $K_a$ signal from the crystalline Si target [8] and thin Cu foil [9].

For excited core-hole -atom or -ion populations, SF has no time to develop due to the fast Auger decay and the pump pulse must be intense enough to build up an inverted population of core-hole excited atoms or ions in time shorter than the corresponding lifetime [10]. While quasiprompt SASE is normally detected in the spectral domain by measuring the nonlinear dependence of the total photon yield on pump intensity, a time resolved measurement is required to study the much slower onset of the SF signal with linear dependence on pump intensity. This may be quite straightforward when singly excited valence states with lifetimes of the order of 1 ns or longer are involved and if the emission energy is not too low; for visible light, the temporal resolution of a streak camera can be as low as 0.2 ps, approaching the duration of FEL pulses [3]. Above 1 $\mu$m emission wavelength the temporal resolution of a streak camera based on a solid-state photocathode drops to several tens of picoseconds. To reach the required picosecond resolution at wavelengths longer than $\sim1.6$ $\mu$m, one instead resorts to the pump-probe transient absorption technique [11] or employs the streak camera arrangement with Rydberg atoms playing the role of a photocathode [12]. For some special systems there have been attempts in the past to indirectly capture both SF and SASE phenomena by controlling the decoherence time of the system, for example by varying the temperature of the O$_2$-doped KCl crystal [13].

This paper deals with collective effects in the most basic $\Lambda$ configuration involving the $1^S$, $2^1P$, and $2^1S$ states of the helium atom (Fig. 1). As this example is devoid of uncertainties related to cascade decays, it can serve as a benchmark case; the theoretical analysis presented in the paper may be complemented by implementation of the IR radiation detection scheme with a picosecond temporal resolution in the near future. To the purpose, the analysis is performed for a broad range of pump intensities ($10^9$–$10^{13}$ W cm$^{-2}$) and target pressures ($10^{-3}$–$10^3$ Pa) with a fully coupled pump and decay scheme on a time scale going well beyond the $2^1S$ lifetime. The evolution of the radiation patterns is therefore followed from

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FIG. 1. Scheme of the lowest singlet energy levels in the helium atom forming a benchmark Λ system.

the weak-pump–low-pressure spontaneous emission regime to the superfluorescence regime, dominating at high pump intensity and target pressure. The 21P state features only one dipole-allowed transition at long wavelengths, so there is no competing SF. Moreover, the upper state is well isolated in the spectrum: the closest dipole-allowed 31P state has 2.1 eV higher excitation energy, much larger than the spectral width of a seeded FEL pump, such as FERMI [14]. Another advantage is that an atom in the 21P excited state is relatively small and therefore less disturbed by atomic collisions. Finally, since the pump wavelength is below the ionization threshold, plasma effects are negligible. Indeed, it turns out that, at the highest pump intensity and target pressure considered here, less than 1% of the ground-state population is ionized by a two-photon resonant absorption involving 21P as an intermediate state.

II. SIMULATION

The corresponding three-level Λ system assumes that the 21P state (i) with τ = 555 ps lifetime in the single-atom limit [15] is resonantly pumped from the helium ground state (0) by a 100 fs laser pulse at 58.4 nm. It is known that fluorescence from the excited 21P to the 21S state (f) at λ = 2059 nm is emitted with the decay branching ratio of Bf = 1.0 × 10−3 [16]. The alternative decay channel of the upper state is 21P−11S. The 21S state in helium has a lifetime of 20 ms [17] and is considered to be stable in this study.

Maxwell-Bloch equations (MBE) are used to simultaneously follow the temporal evolution of the atomic states as well as forward propagation of the pump field F and emitted radiation field E. We assume that the irradiated part of the gas has a cylindrical shape with radius r0 = 25 μm and length l = 1 mm. The off-axis emission is neglected, as well as the effects of counterpropagating fields. These approximations are reasonable because collective effects preferentially develop along the cylinder axis unless the gain length becomes comparable to the radius. Moreover, for the chosen geometry the effects of diffraction are small because the Fresnel number πr02/(λl) ≈ 0.95 [2]. For the selected Λ system, MBE assume the following form (in atomic units):

$$\frac{\partial F}{\partial t} + c \frac{\partial F}{\partial \zeta} = -2\pi i \omega_0 \gamma N \rho_{0i}^*,$$  

$$\frac{\partial E}{\partial t} + c \frac{\partial E}{\partial \zeta} = -2\pi i \omega_0 d N \rho_{fi}^*,$$

Here ρnn denotes a matrix element of the density operator $\hat{\rho}$ and $N = p/(k_B T)$ the density of atoms in the target. The 1S−21P (21P−21S) transition energy is denoted by $\omega_0 = 21.218$ eV ($\omega_0 = 602$ eV), while g = 0.24 a.u. (d = 1.61 a.u.) is the corresponding dipole matrix element [18]. Similar to [5], the Doppler line broadening is accounted for in an approximate way by adding an extra decay rate γD$\omega_0$ to $\rho_{0i}$ and $\rho_{fi}$ coherences, respectively, where $\gamma_D = \sqrt{8 \ln(2) k_B T / (M c^2)} \approx 6.15 \times 10^{-6}$ at room temperature, with M being the mass of the atoms and T the gas temperature. The above equations were rewritten in their finite difference form and numerically propagated on a two-dimensional grid with one spatial dimension [19]. The phenomenological term $S_{fi}$ creates fluctuations of the dipole moment and was added to Eq. (6) to mimic decay due to spontaneous emission. The action of

$$S_{fi} = i \sum_{n,m} A_S \exp(i \phi_m) \delta(z - n \Delta z) \delta(t - m \Delta t),$$

$$A_S = \frac{\Omega_{fi} \Gamma_i \Gamma_f \rho_{ii}(n \Delta z, m \Delta t)}{8\pi^2 N \omega_0 d^2},$$

is to impulsively inject a coherence with a randomly selected phase $\phi_m$ at each point $(n \Delta z, m \Delta t)$ of the grid. Emission solid angle $\Omega = \pi (r_0/\lambda)^2 = 2.0 \times 10^{-3}$ in the forward direction is determined by the dimensions of the active medium. The partial decay width due to the 21P−21S emission is denoted by $\Gamma = B_i \Gamma_f$, where $\Gamma_i = \tau^{-1}$ and $\Gamma_f = \Gamma_f + 2 \gamma_D \omega_0$.

The temporal profile of the incoming FEL pulse is assumed to be fully coherent with a Gaussian time dependence of intensity $I(0, t) = I_0 \exp \left[ -4 \ln 2 (t - t_0)^2 / \tau_0^2 \right]$, exhibiting full width at half maximum (FWHM) pulse duration $\tau_0 = 100$ fs. The time offset is set to $t_0 = 360$ fs, i.e., time $t = t_0$ coincides with the moment the pump hits the target entrance (z = 0) with intensity $I_0$. The spatial intensity profile of the pump is assumed to be independent of the distance from the cylinder axis with a cutoff at $r_0$ (a box-type distribution). Some simulation results are presented also for an axially symmetric Gaussian distribution of pump intensity in the perpendicular plane with the FWHM equal to 2r0 and peak intensity $I_0$.

Since the pump pulse duration is much shorter than the delay and width of SF pulses, and because the structure of pulses needs to be better resolved at shorter delays, it was convenient to divide the time propagation into two parts. In the first part, the length of the time step is $\Delta t = 5$ a.u. ($\approx 0.12$ fs). After 12 ps, well after the passage of the pump...
pulse through the target, the time step is increased to 500 a.u. (≈12 fs) and correspondingly so the spatial grid distance \( \Delta z = c \Delta t \). The time propagation of the MBE was terminated after \( t_1 = 2.4 \) ns, to propagate radiation fields well beyond the spontaneous decay lifetime. Due to the random initiation of the emission process the reported state populations and radiation field profiles are averaged over 20 pump shots unless otherwise stated.

III. RESULTS AND DISCUSSION

Calculations were performed systematically over a broad range of pump intensities and target pressures [Fig. 2(a)]. Several different processes were observed: in the limit of low target pressure and pump intensity, the time profile of the emitted pulse shows an exponential tail with a spontaneous decay time of 555 ps and a very broad phase distribution. At somewhat higher pump intensities and/or target pressures, the number of emitted photons starts to increase exponentially with target pressure (or pump intensity), which is a signature of self-amplified spontaneous emission. The delay of the emitted pulse increases, whereas the phase distribution narrows. Increasing the parameter values even more leads to the regime of superfluorescence with a linear increase in the number of emitted photons and decreasing pulse delay and duration. In this regime Rabi oscillations modify the temporal profile of the emitted pulse, resulting in additional peaks in the temporal profile as well as the phase distribution, indicating that the phase of the field remains approximately constant within a single oscillation, but changes when an intensity minimum is reached.

A. Spontaneous emission

Radiation fields were first calculated at low target pressure and pump intensity, where the emission process is spontaneous and absorption of the pump field follows the Beer-Lambert law [20]. In this limit, the calculated average number of photons emitted until the end of the propagation time \( t_1 \) is expected to be a linear function of the target length, and the temporal profile of the emitted radiation intensity at the target exit (\( z = l \)) should decrease exponentially with the decay time \( \tau \). Both of these properties are seen to emerge in Fig. 3(a) when the signal is averaged over a large number of pulses. Moreover, the number of emitted photons was estimated from the number of atoms in the excited \( ^2\!P \) state immediately after the passage of the pump pulse through the target, i.e., before any considerable spontaneous emission could take place. To the purpose, the calculated number of atoms was multiplied by the product \( B_j \Omega_1/(4\pi) \) of the branching ratio for the \( ^2\!P \rightarrow ^2\!S \) transition and the relative emission solid angle. A good match of thus estimated and the calculated average trend in Fig. 3(a) shows that the \( S_{fi} \) term in Eq. (6) has a proper scaling. From the propagated radiation field \( \mathcal{F} \) also the number of absorbed photons until \( t = t_1 \) was calculated as a function of the target length. The calculated trend agrees well with the number of absorbed pump photons estimated from the Beer-Lambert law, when it is taken into account that the Fourier spectral width of the 100 fs pump pulse is much larger than the natural broadening of the \( ^2\!P \) state, given by \( \Gamma_i \) [Fig. 3(a)]. In the weak limit, the number of emitted photons is expected to be a linear function of both the pump intensity and target pressure. The results of the simulations in Fig. 3(b) agree with that.

Because of the relatively large time step in the second part of time propagation and small natural linewidth of the
selected transition (∼1 μeV), it is difficult to generate a precise spectral line shape in the spontaneous emission limit. Still, the simulated spectral shape shows a sharp peak with long Lorentzian tails and has a FWHM in agreement with the spectral width of the $2^1P - 2^1S$ transition. Also, the number of emitted photons as a function of pump detuning is a Gaussian, approximately equal to the spectral image of the pump pulse, because the latter is much broader than the spectral width of the $2^1P$ state.

Although the number of emitted photons in the weak limit agrees with the expected number, the simulated temporal profile of the emission pulse is not entirely exponential: it shoots up too slow compared to the expected, quasiexponential Gales function—a convolution of a relatively short Gaussian pump pulse and exponential spontaneous decay function ($\tau_0 \ll \tau$). As shown in Appendix, this difference is the very consequence of using the kicking term $S_{fi}$ in Eq. (6) to mimic spontaneous emission.

**B. Self-amplified spontaneous emission**

With increasing target pressure and pump intensity, the intensity of spontaneously emitted radiation builds up until at certain target depth it becomes strong enough to stimulate itself. The number of emitted photons then starts to increase exponentially (Fig. 4), which is characteristic of self-amplified spontaneous emission. To find the onset of the SASE process we estimate the critical amplitude of the electric field, $\epsilon_c$, at which the number of photons per unit time, generated in the thin target layer due to stimulation of spontaneous emission from previous layers, equals the number of photons per unit time emitted spontaneously in the solid angle $\Omega$. The population of atoms in the final state starts to build up according to the rate equation

$$\dot{\rho}_{fi} = \Gamma_r \rho_{ii} + \sigma_i(\omega_i)\upsilon I_\epsilon(t)\rho_{ii},$$

where $\sigma_i(\omega) = \sigma_{fi} \Gamma_{fi}(2\pi)^{-1}/(|\omega - \omega_i|^2 + \Gamma_{fi}^2/4)$ is proportional to $\sigma_{fi} = 4\pi^2\alpha_0 d^2$, the energy integrated photoabsorption cross section from the upper state, and photon flux of the seed field is given by

$$I_\epsilon = |\mathcal{E}_e|^2/(8\pi c \alpha_0).$$

Factor $\upsilon < 1$ sets the effective photoabsorption cross section and depends on the temporal profile of the radiation field and its spectral width relative to the width of the transition (see Ref. [20]). For the exponentially decaying field of spontaneous emission both widths are equal and $\upsilon = e^{-1}/2 \approx 0.18$. When the above critical condition is applied to Eq. (11), it follows that

$$\frac{\Gamma_r \Omega}{4\pi} = \frac{\upsilon d^2 |\mathcal{E}_e|^2}{\Gamma_{fi}}.$$

In our specific case, $|\mathcal{E}_e|^2 \approx 8.5 \times 10^{-22}$ a.u. We assume that the critical intensity of radiation in the forward direction builds up by spontaneous emission from the photoexcited target up to the observed point with intensity given by Eq. (A9) in Appendix. Neglecting thus the contribution of previously emitted stimulated emission to the seed field, an estimate of the critical target length is

$$l_c \approx \frac{2 \epsilon_c^2}{\alpha \rho_i \Omega \upsilon |\mathcal{E}_e|^2} \approx \frac{\Gamma_{fi}}{2\pi \alpha \rho_i \upsilon \mathcal{E}_e N d^2}.$$
population is taken to be constant along the critical target length, a good approximation, as long as the absorption of the pump pulse is negligible, i.e., at low target pressure. Then, according to Ref. [20], the following rate equation applies for the population of the upper state:

$$\dot{\rho}_i = \sigma_0 \omega_{01} \text{Re}[F(t)] \hat{F}(t) \ast (\Gamma_{0i}/2) e^{-\Gamma_{0i}/2} \rho_{00}. \quad (14)$$

The asterisk denotes the convolution, \(F(t)\) is the electric field of the pump pulse at target entrance, and \(\Gamma_{0i} = \Gamma_i + 2\gamma_{i0} \omega_{01}\) is the Doppler broadened transition width. The maximum absorption of the pump pulse is given by \(\sigma_0 \omega_{01} = 8\pi \omega_{01} \Gamma_i / \Gamma_{0i}\), which is true for low target pressures. Taking \(\rho_{00} = 1, \rho_{0i}(0) = 0\) and integrating in time, one sees that the population is proportional to the pump intensity,

$$\rho_i = \frac{4\sqrt{\pi} \omega_{01} \gamma_{G0}^2 \tau_0 g_1^G(\xi)}{\Gamma_{0i} \sqrt{\ln 2}} I_0, \quad (15)$$

where \(g_1^G(\xi) = \sqrt{\pi} \xi e^{\xi^2} \text{erfc}(\xi)\) and \(\xi = \tau_0 \gamma_{0i}/(4\sqrt{\ln 2})\). In our case, the \(g_1^G\) factor accounts for a reduced absorption due to the much broader spectral width of the Gaussian pump pulse with respect to \(\Gamma_{0i}\).

Combining the above results and setting the target length equal to the critical length, \(l = l_c\), we finally arrive at the SASE onset condition for a given target: \(I_0 = S_c\), where

$$S_c = \frac{\ln 2}{\pi^2} \frac{4\sqrt{\pi} \omega_{01} \gamma_{G0}^2 \tau_0}{k_B T \gamma_{G0} \Gamma_{0i}} \approx 3.2 \times 10^{-19} \text{ a.u.} \quad (16)$$

The estimated SASE threshold, \(S_c = 6.1 \times 10^{10} \text{ W Pa cm}^{-2}\), is inversely proportional to the target length and depends on the pump pulse duration in a nonlinear fashion. In the log \(p\) versus log \(I_0\) graph, the SASE onset condition reduces to a line separating the spontaneous emission from the SASE region. Indeed, the position of the demarcation line in Fig. 2(a) matches the border between the two regions as determined from the numerical solution of the MBE for various combinations of pump intensity and target pressure. The validity of the onset condition evidently extends beyond the low pump absorption region because the onset of stimulated emission is dominated by spontaneous emission from the first target layers.

In the SASE regime, the duration of the emitted pulse is approximately constant. When target thickness, or target pressure, is increased, the SASE emission is delayed (see Appendix). The exponential growth of the emission intensity continues until half of the upper state population \(\rho_{01}\) in the target layer is transferred to the final state by stimulated emission. Then, even if the external field is increased, the target layer adds only a constant amount of intensity to the field, and the total intensity again grows linearly with target depth. After this turning point the time delay of the emission pulse starts to decrease, signaling the onset of the superfluorescence regime. Both of these trends are evident in Figs. 4(a) and 4(b).

Contrary to the spontaneous emission case, where the time profile of the emitted pulse resembles a two-dimensional random walk in the complex plane, in the SASE regime the profile obtains the shape of an ellipse (Fig. 5) covering a much narrower range of field phases. In the early stages of amplification, the time profile of the emitted field is still considerably influenced by the random nature of spontaneous emission. As the field is further amplified, the profile becomes smooth, meaning that the random emission onset is no longer directly reflected in the emitted field profile.

Figure 4 shows simulation results up to pump intensity \(I_0 = 10^{11} \text{ W cm}^{-2}\). Above this value, the period of the Rabi oscillations due to the pump field becomes comparable to the duration of the pump pulse. This affects the population transfer to the excited state causing the number of emitted photons to oscillate with the pump intensity. This is also seen in the superfluorescence regime and is further discussed below.

**C. Superfluorescence**

In the superfluorescence regime the total radiated intensity increases proportionally to the target pressure, until the photoabsorption of the pump pulse itself becomes nonlinear (Fig. 4). Moreover, the higher \(I_0\), the faster stimulated emission saturates, as indicated by Eqs. (13) and (15).

The upper part of Fig. 6(a) shows temporal profiles of the emitted radiation in the superfluorescence regime for the box-type spatial intensity profile. In combination with Fig. 4, we can observe signature features of superfluorescence: higher pump intensities (and target pressures) lead to shorter pulse delays and durations. Rabi oscillations, which in this context are often called superfluorescent “ringing”, are present in the time profiles. At pump intensities above \(10^{11} \text{ W cm}^{-2}\), the
FIG. 6. (a) Temporal profiles of the emitted superfluorescence pulses and (b) number of emitted photons at different pump intensities and fixed target pressure after transition through a 1-mm-long target for a box-type and Gaussian spatial intensity profile.

number of emitted photons fluctuates around a constant value [Fig. 6(b)]. Since the Rabi frequency of the $2^1P - 1^1S$ transition increases with increasing pump intensity while the duration of the pump pulse remains fixed, the population transfer to the upper state also oscillates with increasing pump intensity. In the complex plane the emitted field profiles have the shape of an ellipse (Fig. 5), the orientation of which shifts as the field propagates through the target. Rabi oscillations in the time profile cause the shape in the complex plane to become more irregular, with each of the peaks in the time profile exhibiting its own, approximately constant field phase, as can be seen from Fig. 2(c).

Realistic FEL sources typically produce pulses with a spatial intensity profile that is close to an axially symmetric Gaussian with the FWHM equal to $2\rho_0$ and peak intensity $I_0$ [1]. We have done the corresponding volume integration by combining simulation results for the box-type intensity distribution with the same $\rho_0$. Effectively, a smooth Gaussian profile in the lateral plane was sampled at selected intensities and then the properly weighted box-type results at these intensities were summed up. The results of the spatial averaging are shown in Fig. 6. The volume integration smooths out the number of emitted photons as a function of pump intensity. At pump intensities below $I_0 \approx 10^{11}$ W cm$^{-2}$ the number of emitted photons is lower than before the integration, simply because the Gaussian spatial intensity profile has a smaller average intensity than the corresponding box-type profile. Above that intensity, the integration results in a higher number of emitted photons, because, due to the Gaussian tail, some of the weights for the fluctuating box-type results are larger than 1. The volume integration also smooths out the temporal profile of the emitted pulse to some degree, although damped oscillations are still present. Similar oscillations have been experimentally observed before in the Rb two-level atomic system [21] and in emission from sodium Rydberg atoms in a resonator [22].

The present theoretical approach is not reliable if pump intensity and/or target pressure are too high. For the case presented in Fig. 5, the gain length (marked by a dashed line) is much larger than the pump beam diameter (dotted line), and the preferred direction for the evolution of stimulated emission coincides with the propagation direction of the pump pulse. When the gain length becomes comparable to or smaller than the pump beam diameter, the probability for an off-axis stimulated emission becomes high and a one-dimensional description of the problem is not adequate anymore. In the limiting case considered in this paper ($I_0 = 10^{13}$ W cm$^{-2}$, $p = 10^3$ Pa), the gain length is approximately 150 $\mu$m, which is still three times larger than the pump beam diameter.

IV. CONCLUSIONS

We have calculated the intensity of the radiation field emitted by a 1-mm-thick layer of helium gas in the forward direction, upon being hit by an intense and coherent EUV pump pulse tuned to the $1^1S - 2^1P$ transition at 21.218 eV. The results are presented for a range of pump intensities and gas pressures, where directionality of the nonlinear $2^1P - 2^1S$ emission at 0.602 eV is imposed by the pump pulse direction. The evolution of the fluorescence signal is studied, starting with an exponential time decay of spontaneous emission, the SASE regime with a delayed emission and high conversion efficiency due to self-amplified spontaneous emission, discussing finally the formation of a strongly peaked signal at reduced delays with damped Rabi oscillations in the superfluorescence regime. While currently existing FEL sources are able to deliver light suitable for experimental realization of this benchmark case, the observation of the IR fluorescence signal in the forward direction with a picosecond resolution may be required to evaluate the presented results.

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APPENDIX

1. Spontaneous emission

Below we model the temporal dependence of $2^1P-2^1S$ radiation by solving the MBE Eqs. (1)–(8) assuming that a low-pressure gas is irradiated by a short light pulse of low intensity. In the spontaneous emission limit it is enough to study emission from a single target slice of thickness $\Delta z = c/\Delta t$ because the radiation intensity from previously excited slices is so low that stimulated emission is negligible: the total emission from the target is obtained simply by summing up contributions from all target slices.

Since in this case the pulse duration $\tau_0$ is much shorter than the lifetime of the upper state $\tau$, the excitation is considered to be impulsive. The initial population $\rho_{0i}(0)$ of the upper state therefore equals some constant $\rho_i$ that depends on pump intensity and atomic target density. The zero time coincides with the moment of the pump passing the slice and the spontaneous emission from the atom is mimicked by a subsequent kicking of the coherence $\rho_{fi}$ with an amplitude $A_i = C/\sqrt{m} e^{-t_i/\Delta t}$ at regular times $t_{m+1/2} = (m + 1/2)\Delta t, \ m \geq 0$. Here we have already inserted a simple exponentially decaying solution of Eq. (4) for $\rho_{fi}(t)$, obtained by neglecting the terms with $\mathcal{E}, \mathcal{F}$ fields. We proceed by solving Eq. (6), which assumes the form of a Langevin equation, where we again neglect the $\mathcal{E}, \mathcal{F}$ terms. We denote the coherence $\rho_{fi}(t) = \rho_{m}$. Taking into account the initial condition $\rho_{fi}(0) = 0$, the solution at times $t_{m+1}$ is given by

$$
\rho_{m+1} = C/\sqrt{m} \sum_{m=0}^{m} e^{-\Gamma_i t_{m+1}} e^{i\phi_{m+y}-(t_{m+1}-t_m-i\phi_{m+y})} \tau_{m+1}. \tag{A1}
$$

Defining $a = \Gamma_i \Delta t/2$ and $b = \Gamma_f \Delta t/2$, the above equation is written in a more compact form

$$
\rho_{m+1} = C/\sqrt{m} e^{-a-b}(m+1)ib \sum_{m=0}^{m} e^{-m(a-b)+i\phi_{m+y}}. \tag{A2}
$$

One sees that the coherence at time $t$ is “forgetting” previous kicks exponentially with time constant $2\Gamma_f^{-1}$ and that memory is being partially restored by the (slower) exponential rise of the kicking amplitude $A_t$ towards time zero (time constant $2\Gamma^{-1}_t$). For small enough time steps $\Delta t$, such that $a, b \ll 1$, the coherence can be considered constant and equal to Eq. (A1) in the time interval $t_{m+1/2} < t < t_{m+1}$, because the $e^{1kb/2}$ correction factors for the coherence at time interval limits are very close to 1. Then, according to Eq. (2), the generated electric field at the slice exit depends linearly on time:

$$
\mathcal{E}(t') = \kappa \left[ (\rho_{m+1} - \rho_{m}^*) t' + \rho_{m}^* \Delta t \right], \quad 0 < t' < \Delta t. \tag{A3}
$$

Here $t'$ runs from the low to the high time range limit, $t_{m+1/2}$ and $t_{m+1}$, respectively, and $\kappa = -2\pi i \omega_0 dN$. At limiting times the electric field is given by

$$
\mathcal{E}_{m+1/2} = \kappa \Delta t \rho_{m}^*, \tag{A4}
$$

$$
\mathcal{E}_{m+3/2} = \kappa \Delta t \rho_{m+1}^*. \tag{A5}
$$

The previously mentioned correction factor to the polarization would induce a correction factor $(1 + b^2/4)$ to the electric fields in Eq. (A4) and is again safely neglected. The radiation field intensity in the selected time interval then scales as

$$
|\mathcal{E}(t')|^2 = |k|^2 [t'^2|\rho_{m+1} - \rho_m|^2 + 2t' \Delta t \text{Re} \{\rho_m^* (\rho_{m+1} - \rho_m)\}]
$$

$$
+ \Delta t^2 |\rho_m|^2]. \tag{A5}
$$

However, at kicking times above the expression reduces to

$$
|\mathcal{E}_{m+1/2}|^2 = |k|^2 |\Delta t|^2 |\rho_m|^2, \tag{A6}
$$

$$
|\mathcal{E}_{m+3/2}|^2 = |k|^2 |\Delta t|^2 |\rho_{m+1}|^2
$$

and, for $0 \leq t' < \Delta t,$ the absolute square of the electric-field amplitude Eq. (A5) is limited by the above two values.

Our aim is to calculate the temporal profile of the emitted radiation intensity averaged over many pump pulses. According to Eq. (A2), the polarization $\rho_{m+1}$ in a single pulse is generated by an (exponentially damped) random walk in two dimensions, exhibiting $m + 1$ steps. It is then easy to see that the radiation intensity Eq. (A6), averaged over many pump pulses, is given by

$$
|\mathcal{E}_{m+1/2}|^2 = \rho_i |C\kappa|^2 |\Delta t|^2 e^{-(a-b)(m+1)b} \times \frac{1 - e^{-2(a-b)(m+1)}}{1 - e^{-2(a-b)}}. \tag{A7}
$$

Inserting back the time variable one gets

$$
|\mathcal{E}(t')|^2 = \rho_i |C\kappa|^2 |\Delta z/e| e^{-\Gamma_f t/a} \times \frac{1 - e^{-(\Gamma_f - \Gamma_j) t_{m+1}}}{1 - e^{-(\Gamma_f - \Gamma_j) \Delta t}}. \tag{A8}
$$

For a small time step $(\Gamma_i - \Gamma_f) \Delta t \ll 1$, the intensity converges to a smooth function of time,

$$
|\mathcal{E}(t)|^2 = \rho_i |C\kappa|^2 |\Delta z/e| e^{-\Gamma_f t/a} \Gamma_f - \Gamma_i. \tag{A9}
$$

It is clear that at large $t$ the above form has $e^{-\Gamma_f t}$ asymptotic time dependence because $\Gamma_f$ is larger than $\Gamma_i$ by definition. However, when $\Gamma_f = \Gamma_i$, the radiation intensity scales as

$$
|\mathcal{E}(t)|^2 = \rho_i |C\kappa|^2 |\Delta z/e| e^{-\Gamma_f t}. \tag{A10}
$$

Without an extra decoherence (Doppler broadening), the selected noise model does not generate the exponentially decaying emission intensity at large times. Figure 7 shows temporal profiles for different $\Gamma_f/\Gamma_i$ ratios together with the numerical result for our specific case. It is evident that for stronger decoherence the simulated pattern better mimics the prompt exponential decay. The matching gets worse when $\Gamma_f/\Gamma_i \rightarrow 1$ with the maximum of emitted radiation intensity shifting towards larger delays $\eta = \ln[\Gamma_f/\Gamma_i]/(\Gamma_f - \Gamma_i)$. The reason for such a behavior is that in the limit the past kicks are not exponentially damped and, consequentially, the absolute square of the sum in Eq. (A2), averaged over many pump pulses, is given by

$$
|\mathcal{E}(t)|^2 = \rho_i |C\kappa|^2 |\Delta z/e| e^{-\Gamma_f t}. \tag{A11}
$$

Given the noise model, the source term needs to be normalized so as to produce the same number of photons as the target does by spontaneous emission in the forward direction. Initially, there are $\rho_i N \text{Sc} \Delta t$ excited atoms in the target slice of thickness $\Delta z$ and surface area $S$, so the slice spontaneously
emits altogether
\[ n_s = \frac{\Omega}{4\pi} \Gamma_f \rho_i N S \Delta z \] (A11)
photons into solid angle \( \Omega \). On the other hand, the total number of emitted photons produced by the source term is given by
\[ n_c = \frac{1}{\omega} c S \int_0^\infty dt |2E(t)|^2. \] (A12)
Inserting \( \rho_i |C_k|^2 \Delta z / (c \Gamma_f) \) for the time integral of Eq. (A9) or (A10), and taking into account the constants
\[ C = i \frac{\Omega \Gamma_f \Gamma_i}{8\pi^2 N \omega d \Delta z}, \]
\[ \kappa = -2\pi i \omega d N, \] (A13)
one verifies that the number of emitted photons is equal to the expected number of emitted photons, \( n_c = n_s \), when kicking term \( S_{fi} \) is given by Eq. (9).

2. Self-amplified spontaneous emission

Below we investigate a buildup of the radiation emitted by the target for different \( \Gamma_f / \Gamma_i \) ratios. The scattered data points mark an average temporal profile of 20 pump pulses for \( \Gamma_f / \Gamma_i = 7.2 \), that applies in the studied case.

FIG. 7. Temporal dependence of the radiation intensity emitted by the target for different \( \Gamma_f / \Gamma_i \) ratios. The scattered data points mark an average temporal profile of 20 pump pulses for \( \Gamma_f / \Gamma_i = 7.2 \), that applies in the studied case.

Below we investigate a buildup of stimulated emission from the spontaneous radiation field created by the passage of the resonant pump pulse through the target. As above, the spontaneous emission field is generated by the action of a random-phase polarization source in each target slice. We look at how the field components with different phases, emitted at different times and places, amplify while traveling through the target. The target is assumed to be fully transparent to radiation, which is a good approximation as long as \( \rho_{ff} \approx 0 \). We assume that the initial population of \( 2^1P \) states is created promptly, i.e., in the moment of the pump pulse passing through the corresponding target slice. Consequently, the population decays exponentially with rate \( \Gamma_i \), as discussed before. According to Eqs. (2) and (6), at time \( t_m \) with respect to the time of the pulse passing the \( n \)th target slice, the slice radiates the field
\[ E_n(t_m) = \sum_{m'}^{m-1} \left[ B_{nn'} e^{\phi_{nn'}}(t_m) + A_{nn'} e^{i\phi_{nn'}} \right]. \] (A14)
The first term of the sum on the right is field due to stimulated emission and stems from the source term \(-i\hbar \rho_{fi}\) in Eq. (2). The seed is radiation field
\[ E_n^{\text{ext}}(t_m') = \sum_{n'}^{n-1} E_{n'}(t_m'), \] (A15)
that is external to the slice and equals the sum of fields emitted by previously excited slices (\( n' < n \)) at the same local time.
The second sum in Eq. (A14) gives the spontaneous emission field of the \( n \)th target slice, generated by adding to \( \rho_{fi} \) of that slice a random-phase contribution at regular time intervals, as discussed above. The process starts by spontaneous emission from the first target slice \((n=0)\) producing the field

\[
E_0(t_m) = \sum_{m'=0}^{m-1} A_{nm'} e^{i\phi_{nm'}}. \tag{A16}
\]

Using Eqs. (A1) and (A4) from Appendix, one verifies that the setting in Eq. (A14) the corresponding phase factor from the first target slice, generated by adding to \( \rho_{fi} \) of the selected phase component \( \phi_f \) of the field is calculated iteratively by setting in Eq. (A14) the corresponding phase factor \( e^{i\phi_f} \) to 1 and all other phase factors to zero. The time dependence of the total field component with phase \( \phi_f \) at position of the \( n \)th slice is obtained by summing up the contributions from all the slices at positions \( z_{n'} \leq z_n \):

\[
E_n^f(z_n, t_m) = \sum_{n'=0}^{n} E_n^f(t_m).
\]

Finally, the time dependence of the total radiation intensity,

\[
\left\langle \sum_{f} \left| E_n^f(z_n, t_m) \right|^2 \right\rangle = \sum_{f} \left| E_n^f(z_n, t_m) \right|^2, \tag{A18}
\]

is given by an incoherent summation of phase-specific intensities because the cross products with different phases average out when many pulses with the random collection of kicking phases are considered.

Figure 8 demonstrates the evolution of the \( 2^1P-2^1S \) radiation field emitted at the onset of the SASE regime. The total radiation intensity in Eq. (A18) was calculated for a 50-mm-long target chopped into slices of thickness \( \Delta z = 1 \) mm. Helium pressure was 15 Pa and the initial population of the \( 2^1P \) state was \( \rho_i = 8 \times 10^{-4} \), as generated by a 100 fs pump pulse with intensity \( 3 \times 10^9 \) W cm\(^{-2} \). The temporal profile of intensity obtained by this simple model is in qualitative agreement with the result of the numerical solution of a full set of MBE Eqs. (1)–(8), performed at the same pump intensity but higher target pressure (700 mbar) and shorter target length (1 mm); see Fig. 2(b).