

Inner-shell x-ray lasing in x-ray free-electron laser-driven highly ionized plasmas

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We present an efficient time-dependent Maxwell-Bloch transport model for simulating inner-shell x-ray lasing (XRL) in x-ray free-electron laser-driven highly ionized and excited nonlocal thermodynamic equilibrium plasmas. The model selectively retains all coherences between lasing levels together with nonradiative coherences linking adjacent lasing transitions. This model fully accounts for all processes governing population evolution, enabling accurate and efficient simulation of complex atomic systems. Applications to neon and argon gases reveal distinct amplification behaviors across charge states. In neon, multiple lasing lines spanning low to high ionization stages are identified, where low-charge-state transitions are strongly shaped by autoionization and saturation, while high-charge-state transitions exhibit pronounced coherence signatures such as Rabi oscillations and Rabi splitting. In argon, amplification is dominated by a few strong channels, while weaker ones are suppressed by nonradiative coherence effects in Λ -type subsystems. Overall, our results demonstrate that laser field-induced multilevel coherent coupling plays a crucial role in shaping XRL gain and spectra. This framework provides a versatile and predictive tool for designing XRLs with tailored temporal and spectral properties, with potential applications in ultrafast science, nonlinear x-ray optics, and precision spectroscopy.

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I. INTRODUCTION

x-ray free-electron lasers (XFELs) have revolutionized the study of light-matter interactions by providing atomic-scale spatial and femtosecond temporal resolution [1,2]. Their high-peak intensity and outstanding transverse coherence have enabled breakthroughs in ultrafast imaging and probing [3,4]. However, their limited longitudinal coherence, with spectral bandwidths typically $\sim 0.1\%$, hinders applications in precision spectroscopy, nonlinear optics, and coherent control [5–7]. To address this challenge, both beam manipulation techniques (such as seeding and self-seeding) [8–12] and alternative lasing schemes [13–16] have been explored. Among these, atomic x-ray lasers (XRLs) based on population inversion provide a promising pathway toward spectrally narrow, temporally coherent x-ray radiation.

Population-inversion-based atomic lasers were first proposed in the 1960s [17] and experimentally realized in the 1980s [18,19], with diverse pumping schemes enabling lasing from the visible to the extreme ultraviolet [20,21]. Extending photoionization-pumped XRLs to shorter wavelengths

was challenging until the advent of XFELs. Thanks to their extremely high-peak intensities, XFELs enabled the first experimental demonstration of a neon XRL at 849 eV (1.46 nm) via rapid K-shell photoionization [14], producing ultrashort, coherent x-ray pulses with narrow bandwidth and a stable central frequency—key advantages over self-amplified spontaneous emission (SASE) XFELs. Since then, XFEL-driven XRLs have been extended to higher photon energies and complex systems. For example, Yoneda *et al.* realized a hard x-ray laser at 8.27 keV (0.15 nm) in copper foils using both amplified spontaneous emission and seeded schemes [22]. Kroll *et al.* demonstrated chemically selective stimulated $K\alpha$ and seeded $K\beta$ emission in manganese systems [23,24]. Doyle, *et al.* achieved seeded stimulated emission (SSE) at 5.9 keV, lowering the pump intensity threshold by nearly 3 orders of magnitude [25]. Zhang, *et al.* generated intense, phase-stable femtosecond hard x-ray pulse pairs via superfluorescence and SSE, enabling attosecond-scale pump-probe studies [26]. Together, these results establish XFEL-driven XRLs as a powerful platform for spectrally pure and temporally coherent x-ray generation.

These experimental advances have, in turn, stimulated significant theoretical developments. Two main approaches have been pursued: (1) rate-equation models [27–32], which describe population dynamics by tracking the diagonal elements of the density matrix and (2) Maxwell-Bloch (MB) formalisms based on the density-matrix method [33–44]. Using rate equations, Rohringer and London [28] investigated XFEL-pumped inner-shell lasing in neon and demonstrated the feasibility of generating intense, subfemtosecond x-ray

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pulses via transient population inversion. Their model also predicted the possibility of multiwavelength lasing from different charge states. Although widely used to analyze amplified spontaneous emission, this framework treats stimulated emission only at the rate level, without retaining the optical phase and quantum coherence of the medium, thereby missing coherence-driven phenomena. In contrast, the Maxwell-Bloch formalism explicitly accounts for atomic coherence, coupling the density matrix of the medium to the radiation field via the macroscopic polarization term. This model has successfully reproduced both XFEL- and plasma-pumped XRL experiments [23,24,45,46]. However, its computational cost scales quadratically with the number of atomic levels involved, making full simulations of complex multilevel atoms or ions impractical. As a result, density-matrix-based models are usually restricted to simplified few-level systems [47], highlighting the need for approximate or hybrid methods that can efficiently capture both population dynamics and coherence effects in complex atoms.

To overcome these limitations, and building on the work of Rohringer and London [28], we extend the theoretical treatment to multilevel systems while incorporating essential coherence effects. Instead of propagating the full density-matrix elements, we introduce a reduced model that retains only two types of coherences: (1) optical coherences between lasing levels and (2) those associated with nonradiative couplings between the terminal states of adjacent lasing transitions that are connected via a common intermediate level (hereafter referred to as nonradiative coherences). These two coherence types are schematically shown in Fig. 1 (red solid and green dashed arrows, respectively). Here, these nonradiative couplings refer not to direct physical transitions between the terminal states, but to quantum coherences in the density matrix that arise from the common coupling of these states to a shared intermediate state, thereby mediating quantum interference between the corresponding lasing channels. All other coherences are neglected in this reduced model. This truncation is physically motivated, as it maintains the coherences most relevant to stimulated emission and cascade amplification, while significantly reducing computational cost. To simulate the lasing process, we develop a one-dimensional Maxwell-Bloch model. The atomic medium is described using a simplified density-matrix representation that preserves full population dynamics and the essential coherences. The model is coupled with a dedicated Monte Carlo algorithm that computes temporal population evolution, enabling simulations of large multilevel systems while keeping computations tractable. As case studies, we apply this method to simulate XFEL-driven photoionization pumping in two atomic gases: neon, comprising 297 energy levels and 627 x-ray transitions included in the propagation and amplification calculations, and argon, involving 56 202 energy levels and 535 such transitions.

This approach captures the full spatiotemporal dynamics of x-ray generation, propagation, and amplification across multiple lasing channels. Compared with rate-equation models, it explicitly incorporates coherence, while compared with full density-matrix treatments, it offers orders-of-magnitude computational savings. The method thus provides a scalable and physically transparent framework for investigating multi-channel coherent lasing dynamics in complex atomic systems.

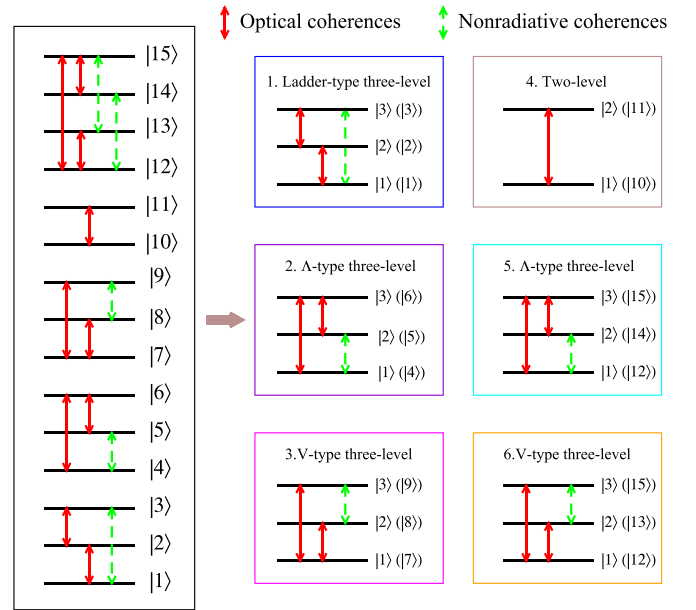


FIG. 1. Schematic illustration of the coherences retained in the reduced Maxwell-Bloch model. The complete set of atomic energy levels (left) is partitioned into constituent lasing subsystems (right), which include ladder-type, Λ -type, and V-type three-level subsystems, as well as two-level subsystems. The selection of subsystems is based on the lasing transitions relevant under the XFEL-driven population-inversion conditions, identified using criteria such as the transition energy and the radiative decay rate. For the Ne example considered in this work, we select lasing transitions whose radiative decay rates exceed 10^9 s^{-1} . Energy levels that do not participate in any selected lasing transition are nevertheless fully included in the rate-equation treatment of population kinetics, represented by brown horizontal dashed lines labeled as $| - \rangle$, where each $| - \rangle$ may denote multiple energy levels. Each level in this diagram represents a fine-structure level characterized by the total angular momentum J , without explicit resolution of magnetic sublevels M . Energy levels included in lasing subsystems are shown as black horizontal solid lines, numbered consecutively for visual clarity. Red solid arrows indicate optical coherences between lasing levels, whereas green dashed arrows indicate nonradiative coherences between terminal states of adjacent lasing transitions. These arrows identify the nonzero off-diagonal density-matrix elements retained in the reduced model; all other coherences are neglected. Notably, subsystems 5 and 6 share the lasing transition $|12\rangle \leftrightarrow |15\rangle$, and many other coupled subsystems of this type can arise. When a lasing transition is shared by multiple three-level subsystems, the time evolution of the corresponding optical coherence (e.g., $\rho_{12,15}$) must incorporate contributions from all subsystems containing that transition. For example, the evolution of $\rho_{12,15}$ is simultaneously driven by nonradiative coherences from subsystem 5 ($\rho_{12,14}$) and subsystem 6 ($\rho_{13,15}$). This subsystem decomposition preserves complete population coupling across the full atomic kinetics through the rate equations, while restricting coherence tracking to localized two- and three-level configurations. This reduces computational complexity while retaining the essential quantum interference effects between coupled lasing channels.

II. THEORETICAL METHOD

The MB formalism is a semiclassical model that describes the coherent interaction between an electromagnetic field and

a quantum system. It couples Maxwell's wave equation for the field propagation with the optical Bloch equations for the quantum-state evolution. A core simplification within the standard MB framework is the rotating-wave approximation (RWA), which neglects the fast-oscillating, nonresonant terms in the interaction Hamiltonian. The RWA significantly simplifies the equations while remaining highly accurate near resonance, making it an essential concept in laser theory and coherent optics. A more general and classical treatment of the MB formalism and the RWA can be found in standard texts, e.g., Ref. [48], with the seminal application of RWA in quantum and semiclassical radiation theories being demonstrated in Ref. [49]. We model the propagation and amplification of XFEL-driven atomic x-ray lasing using a one-dimensional semiclassical MB formalism. The emitted x-ray field is treated as a classical electric field under the slowly varying envelope approximation (SVEA), and its propagation equation is derived from Maxwell's equations coupled to the macroscopic polarization of the medium, which arises specifically from the quantum coherence induced by the lasing transitions. The medium is described quantum mechanically using a reduced density-matrix representation.

The total linearly polarized electric field propagating along the z axis is decomposed into its frequency components, $E(z, t) = \sum_{i \leftrightarrow j} \frac{1}{2} [\mathcal{E}_{ij}(z, t) e^{-i\omega_{ij}t} + \text{c.c.}]$, where ω_{ij} and $\mathcal{E}_{ij}(z, t)$ are the angular frequency and the slowly varying complex envelope associated with the $i \leftrightarrow j$ transition, respectively. The polarization is decomposed analogously, $P(z, t) = \sum_{i \leftrightarrow j} \frac{1}{2} [\mathcal{P}_{ij}(z, t) e^{-i\omega_{ij}t} + \text{c.c.}]$, the polarization envelope is given by $\mathcal{P}_{ij} = 2N\mu_{ij}\rho_{ij}$, where N is the atomic number density, μ_{ij} is the dipole matrix element for the $i \leftrightarrow j$ transition, and ρ_{ij} is the off-diagonal density-matrix element associated with this transition. Under the SVEA, the evolution of the x-ray electric field envelope is given by [33,35,36]

$$\left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) \mathcal{E}_{ij}(z, t) = i \frac{\omega_{ij}}{2\epsilon_0 c} \mathcal{P}_{ij}(z, t), \quad (1)$$

where c is the speed of light in vacuum and ϵ_0 is the vacuum permittivity. We introduce the retarded-time coordinate, $\tau = t - z/c$, which represents the time in the co-moving frame of the propagating x-ray pulse. By transforming the coordinates from (z, t) to (z, τ) , the propagation equation simplifies to $\frac{\partial}{\partial z} \mathcal{E}_{ij}(z, \tau) = i \frac{\omega_{ij}}{2\epsilon_0 c} \mathcal{P}_{ij}(z, \tau)$. This expression highlights that, in the co-moving frame, the field envelope at a given retarded time τ evolves only with propagation distance z .

In the reduced model, only two types of coherences are explicitly retained: (1) optical coherences between lasing levels and (2) nonradiative coherences between terminal states of adjacent lasing transitions (see Fig. 1). Each selected lasing transition is modeled as an independent two-level subsystem when its upper and lower states do not participate in any other selected transition. Whenever two selected transitions share a common atomic level, they are instead grouped into a coupled three-level subsystem to correctly capture population exchange and coherence between the shared state. This subsystem construction follows directly from the selection of lasing transitions, which identifies those most likely to undergo amplification during the propagation of the XRL pulse. Depending on the energy-level structure, the three-level subsystems may be cast into one of three canonical forms: ladder type,

Λ type, or V type. This selective retention allows population transfer through all permitted pathways in the atomic system, while restricting the explicit tracking of coherences to localized two- or three-level subsystems as illustrated in Fig. 1. Coherences between the levels of selected lasing transitions and levels not participating in any lasing transition are typically far off-resonant and decay rapidly due to dephasing, and therefore contribute negligibly to the macroscopic polarization and x-ray gain. While the basic lasing channels are primarily described using simplified two-level or, more commonly, three-level subsystems, the model naturally includes more complex coupled structures. For instance, as indicated in Fig. 1, the coupled levels from $|12\rangle$ to $|15\rangle$ form a network where multiple three-level subsystems share a common transition. Consequently, our reduced Maxwell-Bloch model tracks the full evolution of all populations (diagonal elements of ρ) while retaining only the key coherences identified above. The dynamics of the retained density-matrix elements are governed by the Bloch equations under RWA [47,50].

Population dynamics (diagonal terms). The population dynamics of the density-matrix diagonal elements are given by

$$\begin{aligned} \frac{\partial \rho_{ii}(z, t)}{\partial t} = & \sum_{j \neq i} \rho_{jj} R_{ji} - \rho_{ii} \sum_{j \neq i} R_{ij} \\ & - \sum_{j \neq i} \text{Im}(\mu_{ji} \mathcal{E}_{ji}^* \rho_{ji} / \hbar) + \sum_{j \neq i} \text{Im}(\mu_{ij} \mathcal{E}_{ij}^* \rho_{ij} / \hbar), \end{aligned} \quad (2)$$

where R_{ij} denotes the rate of all incoherent microscopic processes transferring population from level i to j . The last two terms account explicitly for stimulated emission and absorption driven by the coherent x-ray field associated with the retained coherences in the lasing medium.

Coherence dynamics (off-diagonal terms). For each three-level subsystem with states $\{|1\rangle, |2\rangle, |3\rangle\}$, we explicitly retain coherences between lasing levels as well as one additional pair of off-diagonal elements representing nonradiative couplings, as illustrated below.

(1) *Ladder type*

$$\begin{aligned} \frac{\partial \rho_{32}(z, t)}{\partial t} = & -\frac{\Gamma_{32}}{2} \rho_{32} + \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32} (\rho_{33} - \rho_{22}) \\ & + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21}^* \rho_{31} + S_{32}, \end{aligned} \quad (3)$$

$$\begin{aligned} \frac{\partial \rho_{21}(z, t)}{\partial t} = & -\frac{\Gamma_{21}}{2} \rho_{21} + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21} (\rho_{22} - \rho_{11}) \\ & - \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32}^* \rho_{31} + S_{21}, \end{aligned} \quad (4)$$

$$\begin{aligned} \frac{\partial \rho_{31}(z, t)}{\partial t} = & -\frac{\Gamma_{31}}{2} \rho_{31} + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21} \rho_{32} \\ & - \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32} \rho_{21}. \end{aligned} \quad (5)$$

Here, $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |2\rangle$ are lasing transitions, while $|3\rangle \leftrightarrow |1\rangle$ represents a nonradiative coupling between the terminal states of the adjacent lasing transitions. $\Gamma_{ij} = \Gamma_i + \Gamma_j$ is the total decoherence rate for the off-diagonal density-matrix element ρ_{ij} . Here, Γ_i denotes the total decay rate of level i , including both the total Auger decay rate and the

total radiative decay rate. Elastic-collision-induced dephasing, which perturbs the phase of the dipole without causing population decay, is neglected in the present study. This is justified by the relatively low ion densities under our XFEL-driven conditions for Ne and Ar gases, where population-relaxation processes dominate the decoherence dynamics. The term S_{ij} represents a stochastic source modeled as complex Gaussian white noise with variance chosen such that the ensemble average reproduces the correct spontaneous emission rate of the $i \leftrightarrow j$ transition. This stochastic approach was first discussed in Ref. [33], to which Refs. [35,36] refer.

(2) Λ type

$$\frac{\partial \rho_{31}(z, t)}{\partial t} = -\frac{\Gamma_{31}}{2} \rho_{31} + \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} (\rho_{33} - \rho_{11}) - \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32} \rho_{21} + S_{31}, \quad (6)$$

$$\frac{\partial \rho_{32}(z, t)}{\partial t} = -\frac{\Gamma_{32}}{2} \rho_{32} + \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32} (\rho_{33} - \rho_{22}) - \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} \rho_{12} + S_{32}, \quad (7)$$

$$\frac{\partial \rho_{21}(z, t)}{\partial t} = -\frac{\Gamma_{21}}{2} \rho_{21} + \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} \rho_{23} - \frac{i}{2\hbar} \mu_{32} \mathcal{E}_{32}^* \rho_{31}. \quad (8)$$

Here, $|3\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |2\rangle$ are lasing transitions, while $|2\rangle \leftrightarrow |1\rangle$ represents a nonradiative coupling.

(3) V type

$$\frac{\partial \rho_{31}(z, t)}{\partial t} = -\frac{\Gamma_{31}}{2} \rho_{31} + \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} (\rho_{33} - \rho_{11}) + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21} \rho_{32} + S_{31}, \quad (9)$$

$$\frac{\partial \rho_{21}(z, t)}{\partial t} = -\frac{\Gamma_{21}}{2} \rho_{21} + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21} (\rho_{22} - \rho_{11}) + \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} \rho_{23} + S_{21}, \quad (10)$$

$$\frac{\partial \rho_{32}(z, t)}{\partial t} = -\frac{\Gamma_{32}}{2} \rho_{32} + \frac{i}{2\hbar} \mu_{21} \mathcal{E}_{21}^* \rho_{31} - \frac{i}{2\hbar} \mu_{31} \mathcal{E}_{31} \rho_{12}. \quad (11)$$

Here, $|2\rangle \leftrightarrow |1\rangle$ and $|3\rangle \leftrightarrow |1\rangle$ are lasing transitions, while $|3\rangle \leftrightarrow |2\rangle$ represents a nonradiative coupling.

In multilevel systems, single coherence can be shared by several three-level subsystems. If an optical coherence ρ_{ij} is shared across multiple subsystems (e.g., $\rho_{12,15}$ shared by subsystems 5 and 6 in Fig. 1), it is still evolved by a single equation [e.g., Eqs. (3), (4), (6), (7), (9), or (10)], but any driving term must incorporate cumulative contributions from all nonradiative coherences defined in all three-level subsystems that contain the $i \leftrightarrow j$ transition. Similarly, if a nonradiative coherence ρ_{ij} is shared, its evolution equations [Eqs. (5), (8), or (11)] must account for the cumulative influence of all optical coherences of the two lasing transitions within every three-level subsystem in which ρ_{ij} acts as the nonradiative coupling between states i and j . The sharing of optical or nonradiative coherences therefore introduces coherence-level

cross-coupling between three-level subsystems, in addition to the population-level coupling already present.

For comparison, we also consider a two-level approximation that retains only the coherence between the upper and lower lasing states:

$$\frac{\partial \rho_{ji}(z, t)}{\partial t} = -\frac{\Gamma_{ji}}{2} \rho_{ji} + \frac{i}{2\hbar} \mu_{ji} \mathcal{E}_{ji} (\rho_{jj} - \rho_{ii}) + S_{ji}. \quad (12)$$

The XFEL pump is treated as a photon flux, and its intensity evolution is described by

$$\frac{\partial I_v(z, t)}{\partial z} + \frac{1}{c} \frac{\partial I_v(z, t)}{\partial t} = -\mu'_v(z, t) I_v(z, t), \quad (13)$$

where I_v represents the pump-laser intensity at photon energy $h\nu$, and μ'_v is the dynamically evaluated effective absorption coefficient accounting for both absorption and stimulated emission processes.

Equations (1)–(13) are solved self-consistently. The population dynamics [Eq. (2)] are treated using a Monte Carlo algorithm that simulates ionization-relaxation processes over a large atomic ensemble [51]. Monte Carlo approaches for solving large collisional-radiative rate equation systems have been previously discussed, e.g., by Peyrusse [52] and by Son and Santra [53], providing methodological context for their application to XFEL-driven atomic systems. The off-diagonal elements of the density matrix [Eqs. (3)–(11) or (12)] are integrated using a Strang split-step scheme, which separates the decay, field-driven coupling, and source terms to ensure numerical stability and efficiency. The emitted x-ray field [Eq. (1)] and pump-laser intensity [Eq. (13)] are solved using the third-order Adams-Bashforth method. Ensemble averages over multiple realizations account for the stochastic nature of spontaneous emission.

III. RESULTS AND DISCUSSION

In this work, we theoretically investigate the propagation and amplification of x-ray emission arising from the interactions of an XFEL with atomic gases of neon (Ne) and argon (Ar), respectively. The photon energies of the XFEL pulses are set to 1400 eV for Ne and 4450 eV for Ar, both exceeding the ionization energies of their hydrogenlike ions—1362.20 eV for Ne^{9+} and 4426.22 eV for Ar^{17+} [54]. These photon energies are above all K-shell binding energies, ensuring that sequential photoionization can strip the atoms completely to the bare nucleus. Under these conditions, the dominant ionization pathway involves inner-shell photoionization followed by the filling of the resulting core vacancy via autoionization of outer-shell electrons [2]. To fully represent the ionization pathways under XFEL irradiation, we include, for any ion in ionization stage x , all electronic configurations obtained by removing x electrons from the ground configuration of the neutral atom. For example, for Ne^{2+} , starting from the ground configuration of neutral Ne ($1s^2 2s^2 2p^6$), all possible electron configurations consistent with the removal of two electrons from the ground configuration are considered: $1s^2 2s^2 2p^4$, $1s^2 2s^1 2p^5$, $1s^2 2s^0 2p^6$, $1s^1 2s^2 2p^5$, $1s^1 2s^1 2p^6$, and $1s^0 2s^2 2p^6$, yielding six configurations and 17 fine-structure levels. From Ne^{0+} to Ne^{10+} , this procedure produces 63 configurations corresponding to 297 energy levels. Applying the

same selection criteria to $\text{Ar}^{0+} - \text{Ar}^{18+}$ results in 1323 configurations and 56 202 energy levels in total. In the present work, each fine-structure level included above is characterized by its total angular momentum J , without explicitly resolving the magnetic sublevels M . The populations within a given J level are assumed to be equally distributed among its M sublevels. This J -level treatment is justified because the dominant ionization and decay mechanisms— $1s$ photoionization, Auger decay, and radiative decay—do not produce significant alignment. At the photon energies used in this study, $1s$ photoionization dominates and does not generate alignment in the absence of electron-correlation effects [55]. While recent theoretical studies have shown that electronic alignment can be induced by a linearly polarized intense XFEL pulse in atomic argon gas, resulting in nonuniform population among M sublevels when L -shell electrons are initially ionized [56], under the photon energies and pulse parameters used in the present study, $2p$ photoionization is weak and this alignment effect is negligible. Furthermore, Auger decay and radiative decay do not create alignment when the initial state is unaligned. Therefore, any nonuniform population among the M sublevels within a given J level remains negligible, so treating each level at the J -resolved level is adequate for describing the population kinetics relevant to the x-ray lasing gain in this work. Nevertheless, the current framework can be naturally extended to incorporate M -resolved population dynamics and coherences, which will allow future investigations of polarization-dependent features of the emitted x-ray radiation.

In the examples considered in this work, the ion density is relatively low. Consequently, during the interaction of intense XFEL pulses with atomic gases, photoionization induced by the laser field is the dominant channel, while electron-impact ionization, excitation, and recombination contribute only marginally. The produced free electrons can also escape efficiently from the dilute gas, further suppressing collisional effects. A comparison of simulations with and without these electron-impact processes shows only minor differences in the resulting ionic populations. Therefore, under the present low-density conditions considered in this work, such microscopic collisional channels are omitted, and the dominant atomic processes retained in this work are photoionization, autoionization, and spontaneous emission. Direct double autoionization [57,58] is also included because it plays a significant role in the overall ionization dynamics during XFEL interactions [51,59]. Stimulated emission and absorption driven by the emitted x-rays are included for all transitions actively involved in the propagation and amplification process, as described in Eq. (2). All atomic data required for the simulation—including energy levels, photoionization cross sections, autoionization rates, and spontaneous emission rates—are generated using detailed level accounting with the Flexible Atomic Code [60].

A. Propagation and amplification of x-ray emission in XFEL-neon gas interaction

We first discuss the interaction between the XFEL pulse and a Ne atomic gas. The XFEL pulse is assumed to be a Gaussian laser pulse with a peak intensity of $5.5 \times 10^{17} \text{ W/cm}^2$

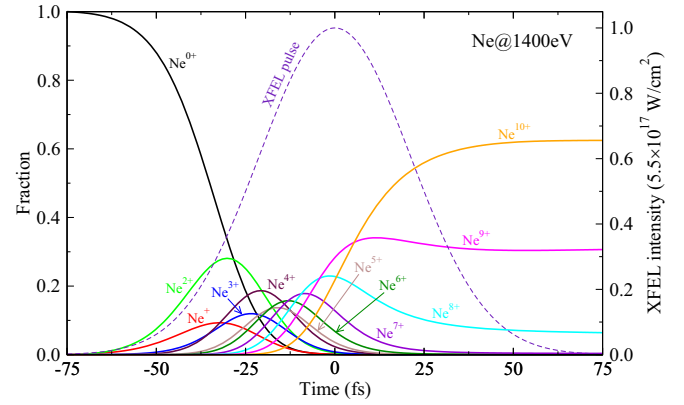


FIG. 2. Temporal evolution of the population fractions of various Ne ionic charge states during the interaction between an XFEL and a Ne atomic gas. The XFEL pulse has a photon energy of 1400 eV, a peak intensity of $5.5 \times 10^{17} \text{ W/cm}^2$, and a Gaussian temporal profile with a pulse duration of 50 fs.

10^{17} W/cm^2 and a duration of 50 fs. Figure 2 shows the temporal evolution of the population fractions of various Ne ion charge states during the interaction of the XFEL pulse with the Ne gas. The populations are obtained by solving rate equations for the local atomic response to the XFEL pump, prior to considering the propagation and amplification of the emitted x-rays. The XFEL pulse irradiates the Ne gas from -75 to $+75$ fs, with the peak intensity reached at $t = 0$ fs. As seen in Fig. 2, the neutral Ne atoms are rapidly ionized, with their population fraction dropping below 10^{-3} by $t = 0.55$ fs. The ions from Ne^+ to Ne^{7+} are rapidly produced but quickly further ionized, resulting in short lifetimes. At the end of the XFEL pulse ($t = 75$ fs), the dominant ionic charge-state distribution is Ne^{7+} (0.38%), Ne^{8+} (6.42%), Ne^{9+} (30.65%), and Ne^{10+} (62.46%).

To theoretically simulate the propagation and amplification of x-ray emission generated in the interaction between an XFEL and a Ne atomic gas, we assume an ion density of the Ne gas of $5 \times 10^{18} \text{ cm}^{-3}$ and a total medium length of 10 mm. This work simultaneously models the propagation and amplification of 627 x-ray transitions with transition energies above 840 eV and spontaneous emission rates greater than 10^9 s^{-1} in Ne ions. These 627 x-ray transitions form 3051 pairs of adjacent transitions (i.e., two transitions sharing a common energy level), which correspond to 3051 three-level subsystems. Many of these 3051 three-level subsystems share common coherences.

Figure 3 shows the theoretically simulated spectral distributions of the 627 x-ray transitions emitted during the interaction of the XFEL with the Ne gas at medium positions $z = 0.1$, $z = 1.57$, and $z = 10.0$ mm. As shown in Fig. 3, when the emitted x-rays propagate to $z = 0.1$ mm, the amplification effect is still negligible, and the spectral distribution is mainly governed by the evolving ion populations under XFEL-Ne interaction and the spontaneous emission rates of the x-ray transitions. Upon propagation to $z = 1.57$ mm, the intensities of several x-ray lines are amplified by several orders of magnitude, especially those emitted from Ne^{8+} and Ne^{9+} ions. After propagation to $z = 10.0$ mm, only a limited

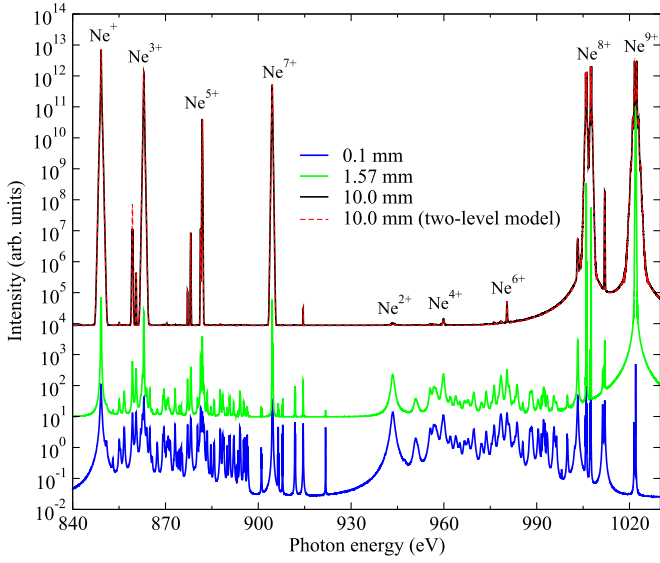


FIG. 3. Spectral distributions of x-ray emission at different propagation distances in the Ne gas medium during the interaction with an XFEL. Solid lines represent results simulated using the three-level approximation, while dashed lines correspond to the two-level approximation. The XFEL pulse has a peak intensity of 5.5×10^{17} W/cm² and a pulse duration of 50 fs. The ion density of the Ne gas is 5×10^{18} cm⁻³, and the total medium length is 10 mm.

number of transitions undergo strong amplification and remain clearly visible in the spectrum, whereas most others experience little to no amplification and remain too weak to be observed. The prominent amplified x-ray transition lines originate from

- (1) Ne^+ : $1s2s^22p^6 (^2S) \rightarrow 1s^22s^22p^5 (^2P_{3/2}^o)$ and $1s2s^22p^6 (^2S) \rightarrow 1s^22s^22p^5 (^2P_{1/2}^o)$,
- (2) Ne^{3+} : $1s2s^22p^4 (^2D_{5/2}) \rightarrow 1s^22s^22p^3 (^2D_{5/2}^o)$, $1s2s^22p^4 (^2D_{5/2}) \rightarrow 1s^22s^22p^3 (^2P_{3/2}^o)$, and $1s2s^22p^4 (^2D_{3/2}) \rightarrow 1s^22s^22p^3 (^2D_{3/2}^o)$,
- (3) Ne^{5+} : $1s2s^22p^2 (^2D_{5/2}) \rightarrow 1s^22s^22p (^2P_{3/2}^o)$, $1s2s(^1S)2p^3 (^2D_{5/2}^o) \rightarrow 1s^22s2p^2 (^2P_{3/2})$, and $1s2p^4 (^2D_{5/2}) \rightarrow 1s^22p^3 (^2D_{5/2}^o)$,
- (4) Ne^{7+} : $1s2p^2 (^2D_{5/2}) \rightarrow 1s^22p (^2P_{3/2}^o)$ and $1s2p^2 (^2D_{3/2}) \rightarrow 1s^22p (^2P_{1/2}^o)$,
- (5) Ne^{8+} : $2s2p (^3P_2^o) \rightarrow 1s2s (^3S)$, $2s2p (^1P_1^o) \rightarrow 1s2s (^1S)$, $2p^2 (^3P_1) \rightarrow 1s2p (^3P_0^o)$, $2p^2 (^3P_2) \rightarrow 1s2p (^3P_1^o)$, $2p^2 (^3P_2) \rightarrow 1s2p (^3P_2^o)$, and $2p^2 (^2D_2) \rightarrow 1s2p (^1P_1^o)$,
- (6) Ne^{9+} : $2p (^2P_{3/2}^o) \rightarrow 1s (^2S)$.

In Ne^{9+} , only the $2p_{3/2} \rightarrow 1s_{1/2}$ transition exhibits strong amplification in our simulations. The $2p_{1/2} \rightarrow 1s_{1/2}$ line contributes negligibly to the observed gain because its population inversion is extremely small under the present XFEL-driven conditions. In principle, microfield-induced Stark mixing could couple the $2s_{1/2}$ and $2p_{1/2}$ levels of H-like neon, whose separation is only the Lamb shift. However, because both levels remain weakly populated under the present XFEL conditions, the resulting redistribution has no appreciable effect on either the population inversion or the x-ray gain. For this reason, neglecting Stark mixing does not significantly affect

the calculated population inversion or x-ray gain, although including these microfield effects would be worthwhile for future extensions of the model at higher plasma densities.

For Ne^+ , Ne^{3+} , Ne^{5+} , and Ne^{7+} , the amplified radiation primarily originates from x-ray emission involving single $1s$ -hole states. In contrast, for Ne^{2+} , Ne^{4+} , and Ne^{6+} , the amplified radiation mainly arises from double $1s$ -hole states, but their amplification is significantly weaker, as is evident from Fig. 3. The limitation originates from the very low population of the corresponding double-hole states. Although sequential XFEL photoionization of single-hole states can produce double-hole states, under the present intensity conditions, most single-hole ions (Ne^+ , Ne^{3+} , and Ne^{5+}) undergo rapid autoionization before a second inner-shell ionization occurs. As a result, although double-hole states are generated during the dynamics, their populations remain too small to establish a strong population inversion. Moreover, the populations of single-core-hole states in even charge ions (e.g., Ne^{2+} , Ne^{4+} , and Ne^{6+}) are also strongly depleted, since most of these ions are produced through fast Auger decay of lower charge states before an additional photoionization can occur, rather than through sequential photoionization that preserves the $1s$ vacancy. Consequently, the resulting Ne^{2+} , Ne^{4+} , and Ne^{6+} ions are predominantly produced without a $1s$ hole. A possible route to mitigate this bottleneck is to enhance the double-hole population by tailoring the excitation conditions. In particular, tuning the XFEL photon energy or combining multiple wavelengths may resonantly access and stabilize these double-core-hole states. Indeed, Mazza, *et al.* have demonstrated that two-color XFEL excitation can efficiently populate $1s^{-2}np$ double-core-hole states in Ne^+ [61], suggesting that similar multicolor strategies could, in principle, be extended to higher charge states to alleviate the present limitation.

For comparison, Fig. 3 also presents the results at $z = 10.0$ mm using the two-level approximation, where only the 627 pairs of off-diagonal density-matrix elements associated with the x-ray transitions are retained. Apart from a distinguishable difference near 859.3 eV, the overall spectral features from the two approximations remain in close agreement. The difference observed near 859.3 eV appears as a variation in the spectral profile of the $1s2s^22p^4 (^2D_{5/2}) \rightarrow 1s^22s^22p^3 (^2P_{3/2}^o)$ transition, which originates from the effects of nonradiative coherences. Certain transitions exhibit significant spectral deviations due to nonradiative coherences, indicating that a three-level treatment is necessary to accurately capture their dynamics. The specific conditions under which nonradiative coherences become essential will be discussed in the following section.

Among the 627 x-ray transitions propagating and amplifying through the medium, eight transitions reach or approach saturation. The evolution of the peak intensities of these XRL pulses as they propagate through the gas medium is shown in Fig. 4. Here, saturation is defined as the intensity at which the rate of stimulated emission equals the total decay rate (including spontaneous emission and autoionization) of the upper lasing state [28]. These include transitions from low-charge-state ions—specifically, the $1s2s^22p^6 (^2S) \rightarrow 1s^22s^22p^5 (^2P_{3/2}^o)$ line of Ne^+ and the $1s2s^22p^4 (^2D_{5/2}) \rightarrow 1s^22s^22p^3 (^2D_{5/2}^o)$ line of Ne^{3+} —as well as transitions from high-charge-

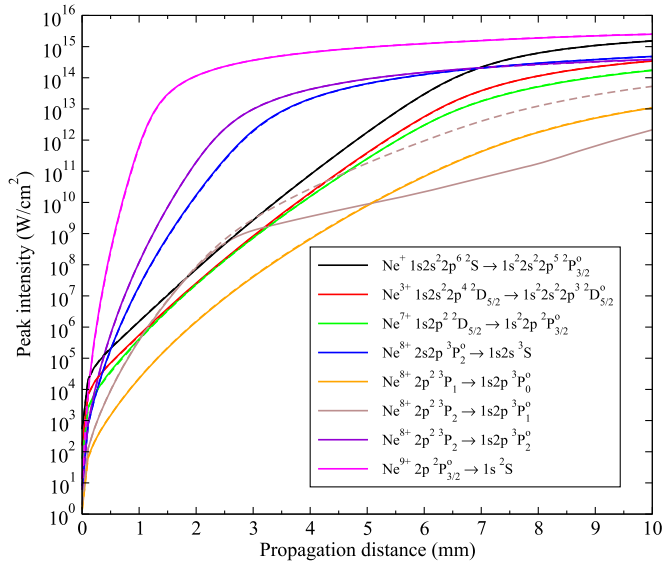


FIG. 4. Evolution of the peak intensities of XRL pulses corresponding to the transitions $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{3/2}^o)$, $1s2s^22p^4(^2D_{5/2}) \rightarrow 1s^22s^22p^3(^2D_{5/2}^o)$, $1s2p^2(^2D_{5/2}) \rightarrow 1s^22p(^2P_{3/2}^o)$, $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$, $2p^2(^3P_1) \rightarrow 1s2p(^3P_0^o)$, $2p^2(^3P_2) \rightarrow 1s2p(^3P_1^o)$, $2p^2(^3P_2) \rightarrow 1s2p(^3P_2^o)$, and $2p(^2P_{3/2}^o) \rightarrow 1s(^2S)$ as functions of the propagation distance of the XRL pulses in the medium. Solid lines represent results from the three-level approximation, while dashed lines correspond to the two-level approximation. Solid and dashed lines of the same color indicate the same transition.

state ions, including the $1s2p^2(^2D_{5/2}) \rightarrow 1s^22p(^2P_{3/2}^o)$ line of Ne^{7+} ; the $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$, $2p^2(^3P_1) \rightarrow 1s2p(^3P_0^o)$, $2p^2(^3P_2) \rightarrow 1s2p(^3P_1^o)$, and $2p^2(^3P_2) \rightarrow 1s2p(^3P_2^o)$ lines of Ne^{8+} ; and the $2p(^2P_{3/2}^o) \rightarrow 1s(^2S)$ line of Ne^{9+} .

For comparison, Fig. 4 also presents the results from the two-level approximation as dashed lines. The peak-intensity evolution curves obtained from the two approximations are in close agreement except for the $2p^2(^3P_2) \rightarrow 1s2p(^3P_1^o)$ transition. All these transitions exhibit rapid exponential growth during the initial propagation stage, followed by a gradual decrease in gain slope as the XRL saturates. Figure 4 shows that the peak intensity evolution of the $2p^2(^3P_2) \rightarrow 1s2p(^3P_1^o)$ XRL pulse simulated with the three-level approximation closely follows that of the two-level approximation during the initial propagation. However, after the XRL pulse reaches the position $z \approx 2.3$ mm, the amplification rate under the three-level approximation decreases noticeably compared to the two-level case. This reduction arises from the effects of nonradiative coherences, which will be analyzed in detail in the following section.

1. Propagation and amplification of x-rays emitted from low-charge-state ions

Due to the substantial differences in the lifetimes of inner-shell excited states between high- and low-charge-state ions, the propagation behavior and spectral characteristics of x-rays emitted from high-charge-state ions can differ markedly from

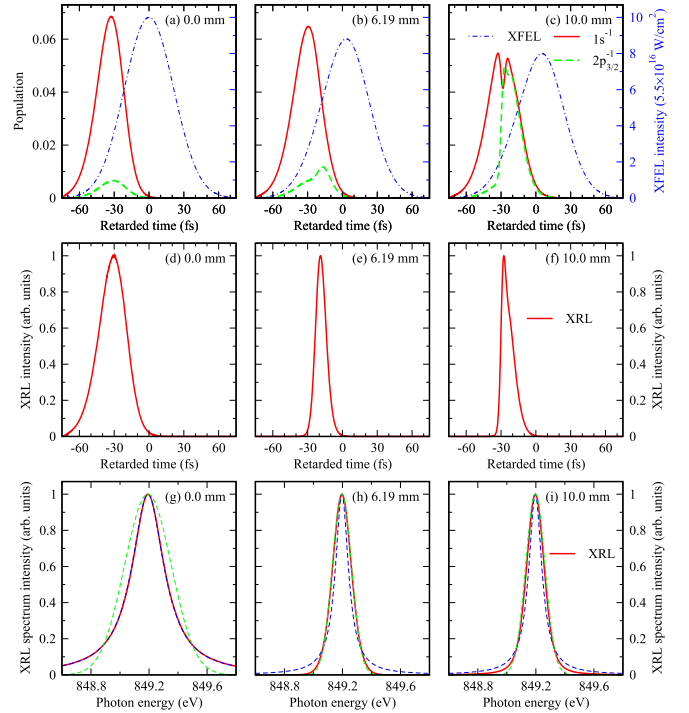


FIG. 5. Time evolution of the populations of the upper level $1s2s^22p^6(^2S)$ and the lower level $1s^22s^22p^5(^2P_{3/2}^o)$ of the radiative transition $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{3/2}^o)$ at positions $z = 0.0$, $z = 6.19$, and $z = 10.0$ mm in the medium, together with the intensity profile of the XFEL pump pulse as a function of time, and the time-domain and frequency-domain spectra of the XRL normalized to the respective maximum values. In panels (g), (h), and (i), the blue dashed lines and green dashed lines represent the Lorentzian and Gaussian line-shape functions, respectively, that best fit the XRL spectral profiles.

those emitted by low-charge-state ions. In the following, we discuss these two categories separately.

As an example of x-ray emission from a low-charge-state ion, we consider the $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{3/2}^o)$ transition line of Ne^+ . Figures 5(a)–5(c) show the temporal evolution of the populations in the upper level $1s2s^22p^6(^2S)$ and the lower level $1s^22s^22p^5(^2P_{3/2}^o)$ along with the time-dependent intensity of the XFEL pump pulse at medium positions $z = 0.0$, $z = 6.19$, and $z = 10.0$ mm. Here, retarded time is used to account for the propagation delay of light in the medium. During propagation, absorption and reshaping reduce the XFEL intensity and shift its peak to later retarded times. At $z = 0.0$ mm, the peak intensity is $5.5 \times 10^{17} \text{ W cm}^2$ at $t = 0.0$ fs. After propagating to $z = 10.0$ mm, it decreases to $4.4 \times 10^{17} \text{ W cm}^2$, with the peak shifted to $t = 5.23$ fs. At $z = 0.0$ mm, the upper-level population reaches a maximum of 6.85% at $t = -32.65$ fs, and the population inversion peaks at 6.2% at $t = -32.9$ fs. With propagation, the growing XRL pulse induces stimulated emission, leading to a gradual reduction of the population inversion. Figures 5(d)–5(f) show the normalized temporal profiles of the emitted XRL pulses at the corresponding medium positions. At $z = 0.0$ mm, the XRL pulse duration (full width at half maximum, FWHM) is 27.6 fs, primarily determined by the upper-level popula-

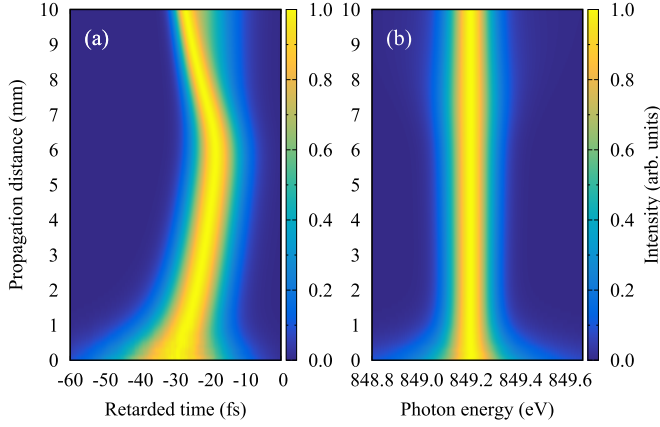


FIG. 6. Normalized (a) temporal and (b) spectral profiles of the XRL pulse for the $1s2s^22p^6 (^2S) \rightarrow 1s^22s^22p^5 (^2P_{3/2}^o)$ transition as a function of propagation distance in Ne gas.

tion. As the pulse propagates, it first narrows due to coherent amplification. At later stages, saturation reduces the population inversion, shifting the maximum amplification to later times and leading to pulse broadening with the formation of a secondary peak. Figures 5(g)–5(i) present the normalized spectral profiles of the emitted XRL pulses. At $z = 0.0$ mm, the spectrum is dominated by spontaneous emission, with Lorentzian and Gaussian fits yield linewidths of 0.27 and 0.38 eV (FWHM), respectively. The Lorentzian value agrees with the measured linewidth of 0.27 eV for the $1s2s^22p^6 (^2S)$ state of Ne^+ [62], confirming that the initial XRL line shape is Lorentzian. As the pulse propagates and amplifies, the spectrum gradually becomes Gaussian-like, but at saturation, secondary peaks appear and the profile departs from a Gaussian form.

Figure 6 shows the evolution of the normalized temporal and spectral profiles of the XRL pulse for the $1s2s^22p^6 (^2S) \rightarrow 1s^22s^22p^5 (^2P_{3/2}^o)$ transition as it propagates through the gas medium. As shown in Fig. 6(a), the retarded time corresponding to the pulse intensity maximum initially shifts to later times, and then shifts back to earlier times. The initial delay is due to absorption and gain dynamics, while the subsequent advancement results from pulse reshaping under local amplification and saturation. The pulse duration decreases rapidly at first and then more gradually, reaching a minimum of 9.1 fs at $z = 8.29$ mm. Beyond this point, secondary peaks appear, gradually broadening the pulse. From Fig. 6(b), the spectral width decreases sharply from the natural linewidth of 0.27 to 0.14 eV at $z = 2.41$ mm. It then increases slightly to 0.16 eV at $z = 7.35$ mm before gradually decreasing again. These temporal and spectral dynamics are consistent with the results reported for $1s2s^22p^6 \rightarrow 1s^22s^22p^5$ of Ne^+ in Ref. [35].

2. Propagation and amplification of x-rays emitted from high-charge-state ions

As an example of x-ray emission from a high-charge-state ion, we consider the $2s2p (^3P_2^o) \rightarrow 1s2s (^3S)$ transition of Ne^{8+} . Figure 7 presents the time evolution of the populations of the upper level $2s2p (^3P_2^o)$ and the lower level $1s2s (^3S)$,

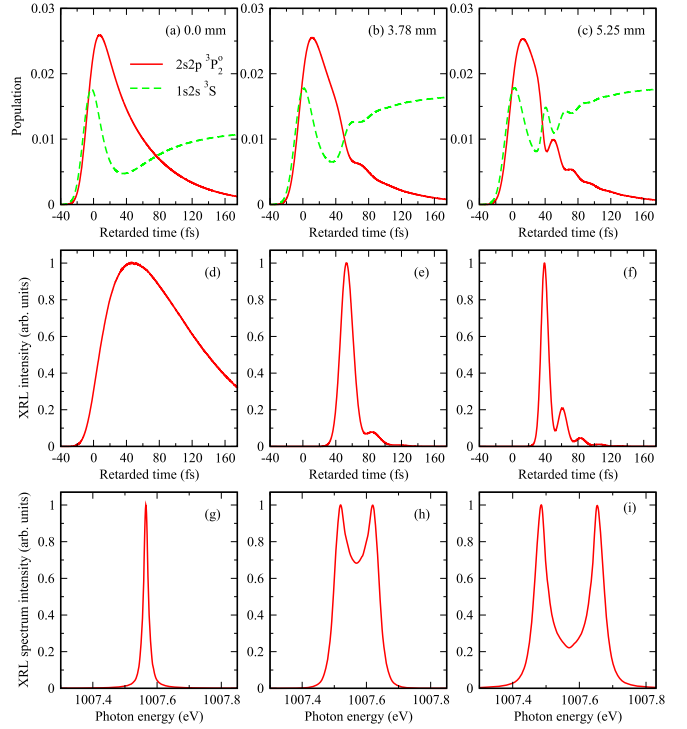


FIG. 7. Time evolution of the populations of the upper level $2s2p (^3P_2^o)$ and the lower level $1s2s (^3S)$ of the radiative transition $2s2p (^3P_2^o) \rightarrow 1s2s (^3S)$, along with the time-domain and frequency-domain spectra of the XRL for this transition (normalized to the maximum value), at positions $z = 0.0$, $z = 3.78$, and $z = 5.25$ mm in the Ne gas medium.

along with the normalized temporal and spectral profiles of the XRL pulse at medium positions $z = 0.0$, $z = 3.78$, and $z = 5.25$ mm. At $z = 0.0$ mm [Fig. 7(a)], the population of the upper level rises rapidly at about $t = -30$ fs, reaches a peak of about 2.6% at about $t = 7$ fs, and then decays gradually. The lower level also increases rapidly, peaking at about 1.8% slightly earlier, and subsequently decreases before slowly recovering. The maximum population inversion reaches approximately 1.6% around $t = 17$ fs. The corresponding XRL temporal profile [Fig. 7(d)] rises from about $t = -32$ fs, peaks near $t = 40$ fs, and has a duration of roughly 130 fs. The spectral profile [Fig. 7(g)] is Lorentzian with a natural linewidth of 0.01 eV. At $z = 3.78$ mm, the upper-level population reaches 2.55% at $t = 11.3$ fs, while the maximum population inversion is 1.45% at $t = 20.35$ fs [Fig. 7(b)]. Stimulated emission drives pronounced Rabi oscillations between the populations of the two levels, producing a secondary peak in the temporal profile of the XRL pulse [Fig. 7(e)] accompanying the dominant amplification peak. The XRL pulse reaches its maximum intensity at $t = 50.05$ fs, with an effective FWHM duration of 18.0 fs determined by the dominant peak. The corresponding spectrum exhibits a Rabi doublet with a 0.1 eV splitting [Fig. 7(h)], a signature of coherent population oscillations. Further propagation to $z = 5.25$ mm enhances the amplification. The Rabi oscillation frequency increases, as evidenced by faster population oscillations [Fig. 7(c)]. The upper-level population peaks at 2.53% at $t = 12.8$ fs, with a maximum population inversion of 1.38%

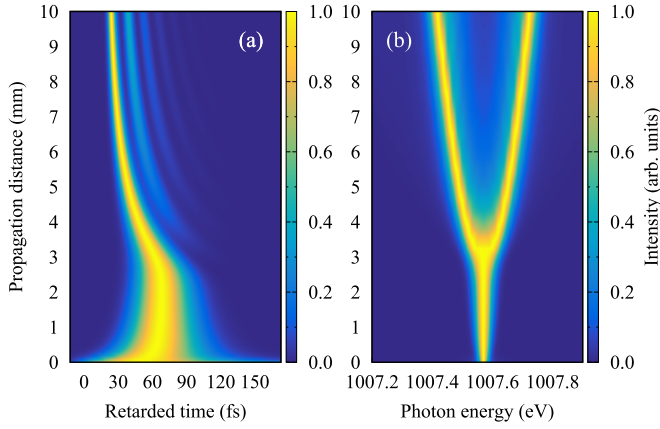


FIG. 8. Evolution of the normalized (a) time-domain spectrum and (b) frequency-domain spectrum of the XRL for the transition $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$ as the XRL pulse propagates through the Ne gas medium.

at $t = 21.35$ fs. The XRL temporal profile [Fig. 7(f)] develops two additional secondary peaks accompanying the main peak, reaches its maximum intensity at $t = 39.2$ fs, and exhibits a further shortened pulse duration of 10.5 fs. The corresponding spectral profile [Fig. 7(i)] shows an increased Rabi splitting of 0.16 eV.

The systematic evolution of the temporal and spectral profiles of the XRL pulse for the $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$ transition is summarized in Fig. 8. As shown in Fig. 8(a), the retarded time of peak intensity initially shifts to later times during propagation, but shifts back to earlier times once strong amplification and pulse reshaping occur. The pulse duration decreases rapidly from ~ 130 fs, then more gradually between $z = 0.5$ mm and $z = 2.5$ mm. Beyond $z \approx 2.9$ mm, secondary peaks emerge in the temporal profile, though the effective duration remains governed by the strongest main peak. The pulse duration continues to decrease, reaching a minimum of 8.5 fs at $z = 10.0$ mm. Figure 8(b) shows the corresponding spectral evolution. The XRL spectrum is initially Lorentzian, determined by the natural linewidth. Coherent amplification gradually makes the profile more Gaussian-like, and further propagation induces broadening and splitting, forming an Rabi doublet at $z = 3.04$ mm. The splitting increases with propagation distance, reaching 0.3 eV at the end of the medium. These features—including Rabi oscillations, spectral splitting, and pulse reshaping—are consistent with previous studies on high-charge-state XRLs under SASE XFEL pumping [36].

3. Effects of nonradiative coherences on x-ray propagation and amplification

Our calculations demonstrate that the gain curves (Fig. 4) and spectral profiles (Fig. 3) obtained within the three-level and two-level approximations can differ substantially. This indicates that coherences associated with nonradiative couplings may strongly influence both the amplification dynamics and the spectral characteristics of the emitted x-rays.

As a representative case, we consider a Λ -type three-level system comprising a common upper state

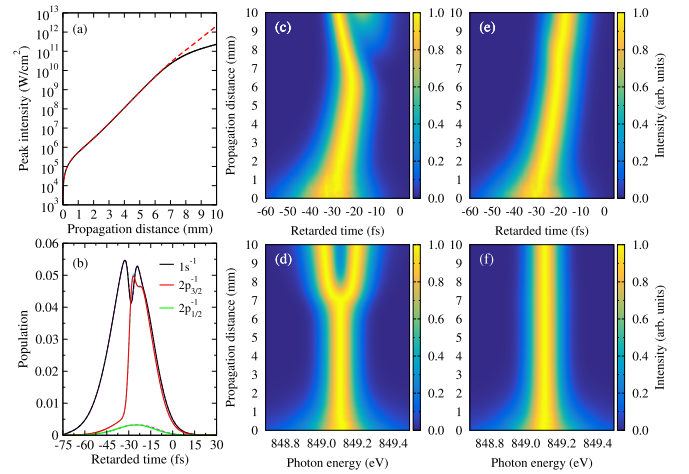


FIG. 9. (a) Evolution of the peak intensities of XRL pulses for the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$ transition in the three-level (solid line) and two-level (dashed line) approximations. (b) Time evolution of the populations of the upper level $1s2s2p6(^2S)$ and two lower levels $1s22s22p5(^2P_{3/2}^o)$ and $1s22s22p5(^2P_{1/2}^o)$ at $z = 10.0$ mm. Normalized temporal (c), (e) and spectral (d), (f) profiles of the XRL pulse for the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$ transition in the three-level (c), (d) and two-level (e), (f) approximations, as functions of the propagation distance in the Ne gas medium.

$1s2s2p6(^2S)$ and the two lower states $1s22s22p5(^2P_{3/2}^o)$ and $1s22s22p5(^2P_{1/2}^o)$, which correspond to the radiative transitions $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{3/2}^o)$ and $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$. We analyze the coherence between the two lower states to illustrate how off-diagonal density-matrix elements associated with nonradiative couplings affect XRL propagation and spectral evolution.

Equations (6) and (7) show that such nonradiative coherences become effective only when the electric field of one of the adjacent radiative transitions becomes sufficiently strong. The field strength of the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$ transition, with a dipole moment of 0.075 a.u. [smaller than 0.106 a.u. for the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{3/2}^o)$ transition], remains relatively small during propagation. Consequently, the coherence between the two lower states exerts only a minor influence on the amplification and spectral profile of the stronger $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{3/2}^o)$ line. In contrast, once the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{3/2}^o)$ transition is sufficiently amplified, the resulting strong field allows the same coherence to significantly alter the amplification rate and spectral profile of the weaker $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$ line. This asymmetric influence, where a strong transition controls a weaker one via a shared coherence, is a clear manifestation of quantum interference in the lasing medium. This coherence-induced coupling is analogous to the interference mechanisms underlying electromagnetically induced transparency [63] and lasing without inversion [64].

This effect is clearly seen in Fig. 9(a): At the early stage of propagation, both the three-level and two-level approximations predict nearly identical gain curves for the $1s2s2p6(^2S) \rightarrow 1s22s22p5(^2P_{1/2}^o)$ XRL. However, once

the intensity of the $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{3/2}^o)$ line reaches $9.59 \times 10^{13} \text{ W cm}^{-2}$ at $z = 6.5 \text{ mm}$, the amplification of the $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{1/2}^o)$ line in the three-level approximation begins to lag behind that in the two-level approximation. At $z = 10.0 \text{ mm}$, the final peak intensities differ by more than an order of magnitude, indicating that coherence between the two lower levels suppresses the amplification of the weaker transition once the stronger line dominates the field dynamics. Figure 9(b) further shows that the population dynamics of the upper and lower levels at $z = 10.0 \text{ mm}$ are nearly identical for both approximations. Therefore, the discrepancy in the calculated gain curves of the $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{1/2}^o)$ transition cannot be ascribed to population differences but instead arises from the coherent coupling between the $1s^22s^22p^5(^2P_{3/2}^o)$ and $1s^22s^22p^5(^2P_{1/2}^o)$ levels induced by the intense laser field.

The role of these coherences is further manifested in the spectral evolution. As shown in Figs. 9(c)–9(f), at short propagation distances both approximations yield nearly identical temporal and spectral profiles. Beyond $z = 6.5 \text{ mm}$, however, the strong field of the $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{3/2}^o)$ transition couples the two lower levels, giving rise to a secondary peak in the temporal pulse profile as well as a quantum interference-induced spectral splitting in the spectrum of the weaker $1s2s^22p^6(^2S) \rightarrow 1s^22s^22p^5(^2P_{1/2}^o)$ transition. Because the photon-energy separation between the two lines is only 0.09 eV and the stronger transition dominates in intensity, the spectral features of the weaker line cannot be resolved in Fig. 3.

The levels $1s2s^22p^4(^2D_{5/2})$, $1s^22s^22p^3(^2D_{5/2}^o)$, and $1s^22s^22p^3(^2P_{3/2}^o)$ form a Λ -type three-level system, with transition energies at 862.99 eV for $1s2s^22p^4(^2D_{5/2}) \rightarrow 1s^22s^22p^3(^2D_{5/2}^o)$ and 859.30 eV for $1s2s^22p^4(^2D_{5/2}) \rightarrow 1s^22s^22p^3(^2P_{3/2}^o)$. In this system, the coherence between the two lower levels modifies the spectral profile of the XRL emission from the $1s2s^22p^4(^2D_{5/2}) \rightarrow 1s^22s^22p^3(^2P_{3/2}^o)$ transition, resulting in a clear difference between the three-level and two-level approximations (Fig. 3). Similarly, the Λ -type system composed of $2p^2(^3P_2)$, $1s2p(^3P_1^o)$, and $1s2p(^3P_2^o)$ exhibits coherence effects between the lower states $1s2p(^3P_1^o)$ and $1s2p(^3P_2^o)$. This coherence leads to observable differences in the XRL gain curve of the $2p^2(^3P_2) \rightarrow 1s2p(^3P_1^o)$ transition when comparing the three-level and two-level approximations (Fig. 4).

B. Propagation and amplification of x-ray emission in XFEL-argon gas interaction

We next investigate the propagation and amplification of x-ray emission induced by an XFEL pulse in an Ar atomic gas. The XFEL pulse has a peak intensity of $1.5 \times 10^{19} \text{ W cm}^{-2}$ and a duration of 30 fs ; the Ar ion density is $1 \times 10^{19} \text{ cm}^{-3}$, and the total gas length is 11 mm . A slightly longer medium (compared to the 10 mm used for neon) is chosen to ensure that several Ar transitions reach full saturation within the simulation domain. Among all possible x-ray transitions, 535 with the strongest population inversion are selected for simulation. These form 1009 pairs of adjacent transitions, corresponding to 1009 three-level systems.

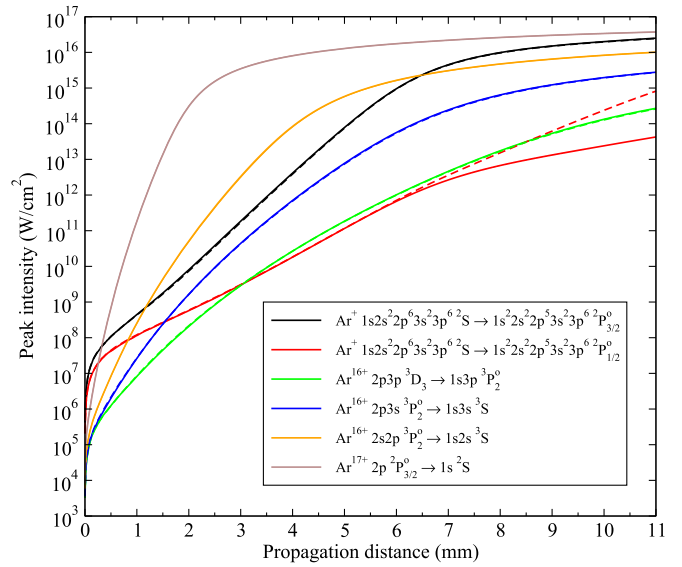


FIG. 10. Evolution of the peak intensities of XRL pulses corresponding to the transitions $1s2s^22p^63s^23p^6(^2S) \rightarrow 1s^22s^22p^53s^23p^6(^2P_{3/2}^o)$, $1s2s^22p^63s^23p^6(^2S) \rightarrow 1s^22s^22p^53s^23p^6(^2P_{1/2}^o)$, $2p3p(^3D_3) \rightarrow 1s3p(^3P_2^o)$, $2p3s(^3P_2^o) \rightarrow 1s3s(^3S)$, $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$, and $2p(^2P_{3/2}^o) \rightarrow 1s(^2S)$ as functions of the propagation distance of the XRL pulses in the medium. Solid lines represent results from the three-level approximation, while dashed lines correspond to the two-level approximation. Solid and dashed lines of the same color indicate the same transition.

After propagation through the medium, only six XRL transitions reach peak intensities above $10^{13} \text{ W cm}^{-2}$ in both the three-level and two-level approximations. These are the $1s2s^22p^63s^23p^6(^2S) \rightarrow 1s^22s^22p^53s^23p^6(^2P_{3/2}^o)$ and $1s2s^22p^63s^23p^6(^2S) \rightarrow 1s^22s^22p^53s^23p^6(^2P_{1/2}^o)$ transitions of Ar^+ ; the $2p3p(^3D_3) \rightarrow 1s3p(^3P_2^o)$, $2p3s(^3P_2^o) \rightarrow 1s3s(^3S)$, and $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$ transitions of Ar^{16+} ; and the $2p(^2P_{3/2}^o) \rightarrow 1s(^2S)$ transition of Ar^{17+} . Figure 10 illustrates the evolution of their peak intensities with propagation distance. For most transitions, the gain curves predicted by the three-level and two-level approximations are nearly identical. A notable exception is the $1s2s^22p^63s^23p^6(^2S) \rightarrow 1s^22s^22p^53s^23p^6(^2P_{1/2}^o)$ line, whose final peak intensity decreases from $8.49 \times 10^{14} \text{ W cm}^{-2}$ (two-level) to $4.33 \times 10^{13} \text{ W cm}^{-2}$ (three-level), which results from nonradiative coherence effects between the levels $1s^22s^22p^53s^23p^6(^2P_{3/2}^o)$ and $1s^22s^22p^53s^23p^6(^2P_{1/2}^o)$.

The spectral evolution of amplified XRL pulses in Ar is qualitatively similar to that in Ne, but high-charge-state transitions exhibit more complex dynamics than low-charge-state ones. Figure 11 presents the normalized temporal and frequency-domain profiles of the $2s2p(^3P_2^o) \rightarrow 1s2s(^3S)$ XRL pulse during propagation. In the time domain, once the pulse reaches sufficient intensity, stimulated emission and absorption induce several secondary peaks. These peaks gradually decay, while shifts in the maximum population inversion generate additional secondary peaks, indicating a continuous reshaping of the pulse profile. In the frequency domain, the spectrum initially splits into a Rabi doublet and subsequently

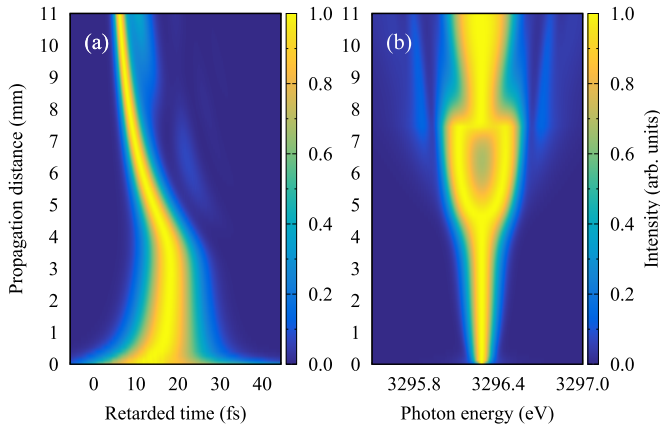


FIG. 11. Evolution of the normalized (a) time-domain spectrum and (b) frequency-domain spectrum of the XRL for the transition $2s2p\ (^3P_2^o) \rightarrow 1s2s\ (^3S)$ as the XRL pulse propagates through the Ar gas medium.

evolves into a dominant main peak accompanied by several weaker secondary spectral peaks. The $2p\ (^2P_{3/2}^o) \rightarrow 1s\ (^2S)$ transition exhibits a similar spectral evolution.

IV. CONCLUSION

We have developed an efficient time-dependent transport simulation method, based on a reduced Maxwell-Bloch model, to investigate the amplification of atomic XRLs driven by XFELs in neon (Ne) and argon (Ar) gases. By selectively retaining all coherences between lasing levels together with the nonradiative coherences linking adjacent lasing transitions, our method achieves computational efficiency and scalability while faithfully capturing the essential physics of XFEL-atom interaction and population evolution. This approach effectively bridges between rate-equation descriptions and full density-matrix treatments, offering a scalable tool for modeling XRL amplification in complex atomic systems.

Our simulations provide detailed insights into the dynamics of highly charged ions in atomic gases under XFEL irradiation. In Ne, multiple lasing lines spanning from low to high charge states were identified. Low-charge-state lines are significantly affected by autoionization and saturation effects, while high-charge-state lines exhibit more pronounced coherence-driven features such as Rabi oscillations and spec-

tral splitting. For double-core-hole states, we found that their limited populations severely limit amplification, suggesting strategies such as multicolor XFEL driving to resonantly enhance and stabilize these states. In Ar, amplification dynamics are dominated by several strong lasing channels, where certain high-charge-state transitions display more complex spectral evolution features than those in Ne. These results reveal possible control mechanisms of XRL output through careful tuning of XFEL parameters and charge-state dynamics.

A key finding is the critical role of nonradiative coherences in multilevel systems. These coherences not only modify the gain by suppressing specific transitions, but also give rise to quantum interference-induced spectral features that are entirely absent in two-level approximations. Such effects clearly demonstrate the limitations of two-level models and underscore the necessity of our reduced density-matrix approach for capturing the full dynamics. This highlights the practical importance of including such coherences for accurate predictions of XRL amplification.

Overall, our work demonstrates a versatile and physically transparent simulation framework, explains the underlying ionization and lasing dynamics across charge states, and reveals the critical impact of multilevel coherences on XRL amplification. Beyond Ne and Ar gases, the method can be extended to three-dimensional propagation, higher-density media, and even solid targets, thus providing a scalable tool for designing XRLs with tailored spectral and temporal properties for frontier applications in ultrafast science, nonlinear x-ray optics, and precision spectroscopy.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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