

Amplification of Spontaneous Emission from Doubly Excited He Atoms

J. Turnšek^{1,2}, Š. Krušič¹, A. Mihelič^{1,2}, K. Bučar^{1,2}, L. Foglia³, R. Mincigrucci³, M. Krstulović³, M. Coreno³, G. Bonano³, K. C. Prince^{3,4}, C. Callegari³, A. Simoncig³, Z. Ebrahimpour³, E. Paltanin³, A. Benediktovitch⁵, R. Osellame⁶, A. G. Ciriolo⁶, R. Martínez Vázquez⁶, C. Vozzi⁶, E. Principi³, and M. Žitnik^{1,2}

¹*Jožef Stefan Institute, Jamova cesta 39, SI-1000 Ljubljana, Slovenia*

²*Faculty of Mathematics and Physics, University of Ljubljana, Jadranska 31, SI-1000 Ljubljana, Slovenia*

³*Elettra-Sincrotrone Trieste S.C.p.A, S.S. 14, I-34149, Basovizza (TS), Italy*

⁴*Department of Surface and Plasma Science, Charles University, Prague, Czech Republic*

⁵*Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany*

⁶*National Research Council (CNR)—Institute for Photonics and Nanotechnologies, Milano, Italy*



(Received 14 January 2025; revised 16 May 2025; accepted 24 July 2025; published 26 August 2025)

We have observed amplification of weak fluorescence from He atoms in the $3a\ ^1P^o$ autoionizing state, which decay by spontaneous emission of 40.74 eV photons in one out of 1600 cases. On average, a $4.1 \pm 1.6\%$ conversion of 63.67 eV photons was detected in the forward direction at 20 μ J pump pulse energy and 40 mbar He gas pressure. A comparison with state-of-the-art theoretical simulations shows an unexpected spectral line shift at high gas pressure that is ascribed to transient molecular and electron scattering effects.

DOI: [10.1103/mg5s-xvct](https://doi.org/10.1103/mg5s-xvct)

Strong excitation of a densely packed atomic ensemble may trigger collective nonlinear response in the fluorescence decay channel, such as amplification of spontaneous emission (ASE) or superfluorescence [1]. In the optical region, these effects have been extensively studied and are a backbone of modern quantum technologies. Promising to boost light intensity, as well as temporal and spatial selectivity, there are persisting efforts to transfer laser techniques toward shorter wavelengths promising to open new research and technological avenues in quantum optics, chemical dynamics, imaging, spectroscopy, interferometry, and lithography.

In the past, many ionic systems were shown to undergo ASE at the extreme ultraviolet (XUV) wavelengths when pumped by the optical-field ionization or fast electric discharge leading to inverted populations via different collisional and recombination schemes [2]. These experiments demonstrate that ASE may deliver radiation down to the water window wavelength range (2.2–4.4 nm) with gain and emission delay depending on properties and evolution of initially prepared plasma. On the other hand, the construction of free electron laser (FEL) facilities [3–6] enables prompt, selective, and efficient excitation of short-lived upper states via single photon absorption. By now,

many systems have been shown to support ASE at short wavelengths when subjected to an intense femtosecond FEL light pulse. This has been achieved in the x-ray region for K-hole excited targets ranging from neon to copper [7–9], showing also that quick buildup of inverted population with a FEL pump in combination with a prompt ASE response preserves spectral line shift due to the initial chemical environment of emitters [9], and that ASE can be selectively enhanced by overlapping the pump with a seed of the ASE-matching wavelength [8,10,11]. These studies are motivated by the possibility to significantly amplify and collimate weak secondary valence-to-core emission, which promises orders of magnitude more efficient x-ray spectroscopy with enhanced chemical sensitivity and excited-state control [12–14]. The self-seeding ASE systems are also in the core of the multipass devices fed by a sequence of FEL pump pulses that have been proposed recently to generate intense x-ray pulses with improved coherence and temporal properties [15–17].

FEL light pulses were also employed to generate inverted populations of atoms or multiply charged ions in singly excited states (SEs), i.e., below the respective ionization thresholds. A dominant decay mode of these upper states is fluorescence in the visible (VIS) or XUV range, which may develop to the collective emission on the picoseconds timescale or longer under proper target conditions [18–22]. On the other hand, the ASE from states above the ionization threshold can be strongly hindered by much faster autoionization or Auger decay, which makes generation of a significant upper state population quite challenging. Moreover, ASE from inner valence holes can

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

be blocked by valence photoelectron and Auger electron collision-driven upper state destruction if the distance between atoms is too small as in solids [23,24].

Below we report on ASE emitted by gas of He atoms in a doubly excited state (DES) below the second ionization threshold ($N = 2$). The selected state quickly autoionizes to the ground state of He^+ ion but can also decay spontaneously by emitting a 30.4 nm photon with a tiny probability of 0.06%. This Letter, therefore, represents a link between the two ASE research lines: the one in the x-ray domain involving core-hole states and the other one dealing with singly excited states below the first ionization threshold ($N = 1$). A relative simplicity of the atomic system and well-defined experimental setup enable detailed comparison of these benchmark results with theoretical predictions for He in order to test and improve our current understanding of the phenomenon. Such studies are of general importance because they expose demands on the spatiotemporal distribution of the upper state population required to support ASE regardless of the pumping method. The acquired knowledge may be useful to handle similar systems, for example, helium-like Li^+ ions in the $2s2p$ state [25,26] that spontaneously emit weak light close to 13.5 nm, the technologically relevant XUV wavelength [27].

At the turn of the millennium it was realized that He atoms in the DES may relax also by spontaneous emission of an XUV photon to populate the SES [28–30]. In such a radiative transition, simply denoted by $Nlnl' \rightarrow 1snl'$, an electron in Nl orbital jumps to an empty $1s$ orbital while the electron in the outer nl' orbital most often remains a spectator [31]. Since Coulomb potential of the He nucleus is relatively weak, DESs are not properly described by a single atomic configuration: for the members of the $na\ ^1P^o$ series of DESs [32] converging to $N = 2$ ionization threshold (yielding He^+ ion in $2s$ or $2p$ state), many-body interactions increase the probability for two electrons to approach each other, as shown by the strong configuration mixing. Correlations in these states, denoted also by $n^+ \approx (2pns + 2snp)/\sqrt{2}$ [33], increase not only the corresponding single photon absorption cross section from the ground state [34] but also the autoionization decay rate, which is much larger than the spontaneous emission rate for low- n members of the series.

To trigger ASE, spectrally isolated $3a\ ^1P^o$ resonance was selected because its lifetime (80 fs) permits efficient preparation of inverted population with 50 fs light pulses from the seeded FEL light source FERMI in Trieste (Italy). In addition, the $na\ ^1P^o$ DES series exhibits the highest spontaneous decay rate with preferential emission of XUV light along the pump pulse direction [31], which helps to start the ASE process. A significant ASE yield from this autoionizing resonance is predicted also by recently published paraxial solutions of Maxwell-Bloch equations with newly derived stochastic noise terms [35] to mimic the

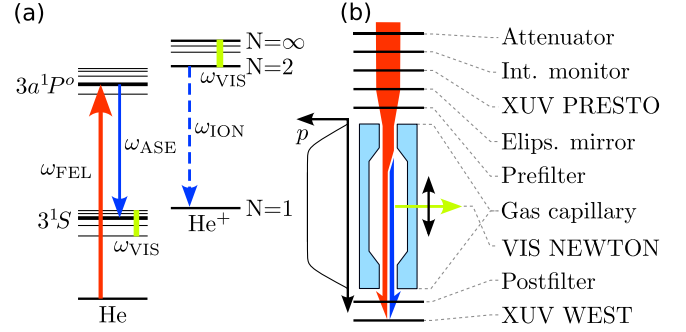


FIG. 1. (a) Electronic transition scheme in He. ω_{FEL} photons (red arrow) excite atoms to the $3a\ ^1P^o$ DES at 63.66 eV. The autoionizing resonance also decays to the $3\ ^1S$ SES by emission of ω_{ASE} photon at 40.74 eV (blue arrow). It is energetically close to $2p \rightarrow 1s$ emission in the He^+ ion at 40.81 eV, denoted by ω_{ION} (dashed blue arrow). Visible fluorescence ω_{VIS} from He and He^+ is denoted by green stripes. (b) Scheme of the experimental setup with the calculated gas pressure distribution in the capillary (see text for description).

spontaneous emission [36]. Note that the widespread modeling with phenomenological impulsive random coherence generators is not suitable to describe ASE initialization in our case because it does not correctly reproduce the temporal profile of the radiation in the spontaneous emission limit [37,38].

Figure 1(a) shows the scheme of atomic and ionic levels playing the role in our ASE experiment on He. The intense XUV pump pulses with 7×10^{-4} spectral bandwidth were tuned to the resonant photon energy $\omega_{\text{FEL}} = 63.66$ eV to maximize the population transfer to the $3a\ ^1P^o$ DES. This state is coupled to the $1s\epsilon p$ continuum with a 3×10^{-4} a.u. transition rate, but also decays radiatively with a much smaller 1.8×10^{-7} transition rate to populate $n(^1S, ^1D)$ SES [39]. We have observed ASE on the $3a\ ^1P^o \rightarrow 3\ ^1S$ transition in He atoms, which results in the emission of ω_{ASE} photons at 40.74 eV. With a 77% branching ratio, this is the most probable radiative decay path of the $3a\ ^1P^o$ resonance in an isolated atom [40]. In parallel to XUV emission, we have measured spectra of VIS fluorescence ω_{VIS} originating from transitions between SES of He and He^+ at later times, in the gas cooling period.

The experimental setup at the EIS-TIMEX beam line at FERMI is schematically presented in Fig. 1(b). Production of ASE was examined for a range of pump pulse energies set by absorption of the surplus light in the nitrogen-filled gas attenuator [41]. The ionization cell monitored transmitted pump energy, and the ellipsoidal mirror focused horizontally polarized light to the He gas target. The two in-line XUV spectrometers, one placed before (PRESTO [42]) and one after the gas target (WEST [43]), measured spectra of light pulses passing at a 50 Hz repetition rate. The VIS spectrometer (NEWTON) was fed by an optical fiber and analyzed light from the gas emitted perpendicular to the

pump pulse propagation. The ASE light, exiting the target in the forward direction, was attenuated by the Ni filter and analyzed by the WEST spectrometer in the second-order Bragg reflection.

The gas cell was micromachined from a block of fused silica glass by removing the material following the cylindrical symmetry [44]. The first and the last millimeter of an open-end microchannel were produced with 100 μm diameter while the 7.2 mm long central portion was made with a diameter of 300 μm . The glass cell was precisely aligned with the focused pump beam by a motorized five-axis manipulator. During the measurements, the glass cell was continuously supplied with He from an external gas reservoir through a wide slit micromachined along its central channel. That way the gas density in the central portion of the cell was practically constant with a rapid drop at both ends. This behavior was confirmed by COMSOL Multiphysics® simulations [45] and by an auxiliary synchrotron experiment verifying an exponential decrease of $3^1P \rightarrow 2^1S$ emission intensity along the gas cell, a clear sign of light attenuation in a homogeneous medium. Under operation, the background pressure in the experimental chamber was just below 10^{-4} mbar with a primary 1350 l/s turbomolecular pump running to maintain 112 mbar of He target pressure, the highest target pressure used in this ASE study.

The well-defined experimental setup enabled comparison of the measured data with theoretically predicted ASE on an absolute scale. The pump pulse energy I_0 at the entrance of the gas capillary was for a factor of 0.65 lower than measured energy in the ionization cell due to the beam transport losses. The transmission of Ni filters for ASE light was determined experimentally by comparing the average ASE yields collected with and without the filters at the same experimental conditions, specified by $\bar{I}_0$, the average pump pulse energy at the cell entrance, and p , the He target pressure. The number N_{ASE} of ASE photons at the capillary exit was estimated with an overall error of 23% by calculating first the number of photons hitting the WEST CCD from integral signal counts on the camera and propagating the light backward by scaling up the photon number with correction factors describing spectrometer geometric losses, spectrometer grating efficiency, and filter transmission [46].

Before comparison with theory, the measured dataset was filtered, i.e., the data collected with pump pulses exhibiting irregular spectra were ignored. The quality of each pulse was judged by its deviation from a Gaussian spectral shape, by the pulse energy and by the central wavelength. The theoretical ASE response was generated by methodology [35], using the calculated longitudinal pressure profile of He gas [Fig. 1(b)] and the set of Gaussian pump pulses closely mimicking the filtered set. The calculations were run assuming Gaussian intensity distribution of the pump in the transverse plane derived

from measured $25 \times 16 \mu\text{m}^2$ FWHM beam size at the cell center. A detailed account on data correction and filtering procedures is given in Supplemental Material [47].

The observed ASE yield shows an exponential increase that saturates upon further increase of the pump pulse energy [35]. In Fig. 2(a), the total ASE yield dependence on pump pulse energy is presented together with the corresponding theoretical result at 40 mbar He pressure. This particular experimental dataset shows an increase of 4 orders of magnitude in the ASE yield as a result of 1 order of magnitude increase of the pump pulse energy. The theory exhibits a similar trend, but the calculated average ASE yield is too low, especially at low pump pulse energies. At least partially, this difference could originate in a departure of the actual pump beam from a Gaussian.

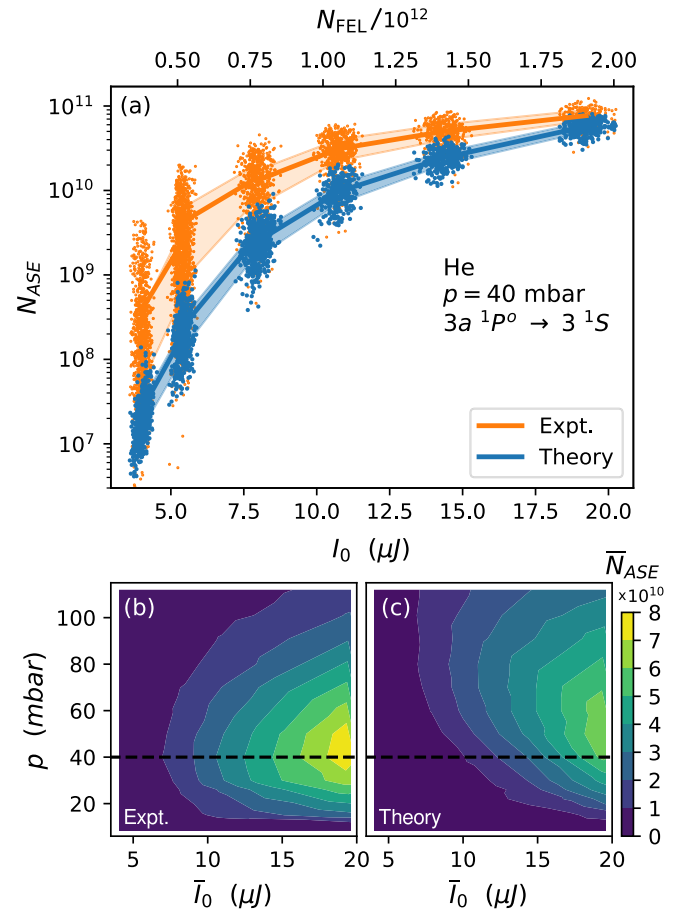


FIG. 2. (a) The number of ASE photons N_{ASE} exiting the target at 40 mbar He pressure at different pump pulse energies I_0 . N_{FEL} denotes the corresponding number of pump photons entering the target. Dots show the experimental (orange) and theoretical (blue) pulse-by-pulse data. Thick curves connect average photon numbers $\bar{N}_{\text{ASE}}$ of data clouds. The interval of N_{ASE} denoted by the shaded area is centered at the corresponding median and contains 50% of accepted pulses. (b) Experimental and (c) theoretical map of the average ASE photon number $\bar{N}_{\text{ASE}}$ at He pressure p and average pump pulse energy $\bar{I}_0$. The dashed line denotes the data cut presented in (a).

The average ASE production at different experimental conditions was examined by sweeping the average pump pulse energy and He pressure in the $2.3 \leq \bar{I}_0 \leq 19.2 \mu\text{J}$ and $8 \leq p \leq 112 \text{ mbar}$ range, respectively. Below these limits, no ASE was detected, and the high limits were imposed by the highest achievable pump pulse energy and gas pumping efficiency. Figure 2(b) shows two-dimensional distribution of the observed average ASE yield, extracted from the measurements at 130 different $(\bar{I}_0, p)$ experimental points gathered at the central pump photon energy ω'_R , where the highest ASE yield was detected. At around 40 mbar, the ridge with a maximum ASE yield is observed that extends from low to high pump pulse energies. At higher pressures the yield diminishes because the gas is becoming optically thick for ASE. The highest measured average number of ASE photons $\bar{N}_{\text{ASE}}$ is $(7.7 \pm 2.9) \times 10^{10}$ at $\bar{I}_0 = (19.2 \pm 0.6) \mu\text{J}$. Defining the conversion efficiency of the ASE process as a quotient between the number of outgoing ASE photons versus the number of incoming pump photons, the highest observed average conversion efficiency is $(4.1 \pm 1.6)\%$. The highest observed single pulse conversion efficiency is $(6.5 \pm 1.5)\%$ and was measured at 40 mbar He pressure and $I_0 = (19.3 \pm 0.2) \mu\text{J}$ pulse pump energy. A comparison with an estimated conversion efficiency of the spontaneous emission ($3 \times 10^{-8}\%$) into the ASE solid angle ($1.6 \times 10^{-5} \text{ srad}$) clearly shows the power of ASE in collimating and amplifying a weak XUV emission signal [40]. While the magnitude and position of the maximum theoretical ASE yield in Fig. 2(c) agree quite well with the experimental data, the theory predicts too high signal at the highest examined values of $\bar{I}_0$ and p . This discrepancy may originate in the absence of transient molecular and inelastic photoelectron scattering effects in the model, which tend to obstruct generation and speed up decay of the DES population. Despite the lower fluorescence branching ratio (0.06% vs 1.8%), the observed ASE conversion in He is higher than 0.1% reported for the Ne K-L line (849 eV) in the seminal work [7], where the population of short-lived $\text{Ne}^+ 1s^{-1}$ states (2.4 fs) was prepared by the 40 fs FEL light pulse tightly focused on the high-pressure Ne gas (660 mbar).

The theory generates pulse-by-pulse data allowing for a comparison with the measured ASE photon number distribution. In Fig. 3, the histograms at 40 mbar He pressure and 19.2 μJ average pump pulse energy are presented. Although the calculations give about 30% less ASE photons for the set of pump pulses mimicking the experimental one, both distributions have approximately the same relative width. The statistical tests do not show significant departures of N_{ASE} histogram from normal distribution in both cases [47–50].

The collected ASE spectra are compared with the theory in Fig. 4. While the calculated ASE yields are too low at low $\bar{I}_0$ (see Fig. 2), the comparison in absolute terms, also

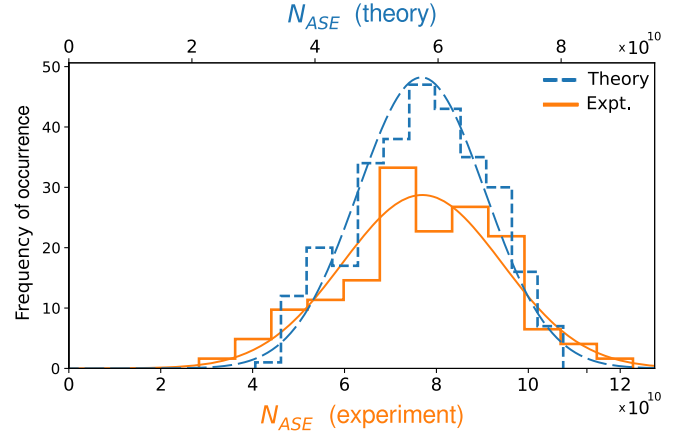


FIG. 3. Experimental and theoretical ASE histograms at $p = 40 \text{ mbar}$ and $\bar{I}_0 = 19.2 \mu\text{J}$. The horizontal scales are adjusted separately to match the most frequently occurring number of ASE photons, and both histograms are normalized to the same area. The vertical scale applies to the experimental data. The curves denote normal distribution fits.

regarding the spectral shape (broadened by the WEST instrumental function), becomes much better at higher pump pulse energies. In addition, we observe a significant pressure-dependent shift of the ASE spectrum, not predicted by simulations at given experimental conditions [35]. The shift may originate from the shift and broadening of the energy levels of the upper and lower states due to the interaction of the emitter with nearby He atoms.

Finally, the pump photon energy scan across the resonance reveals a Gaussian-like dependence of ASE yield [Fig. 5(a)]. Because of the nonlinearity of this process, the observed FWHM (19 meV) is smaller than spectral width of the pump (45 meV) and larger than the natural width of

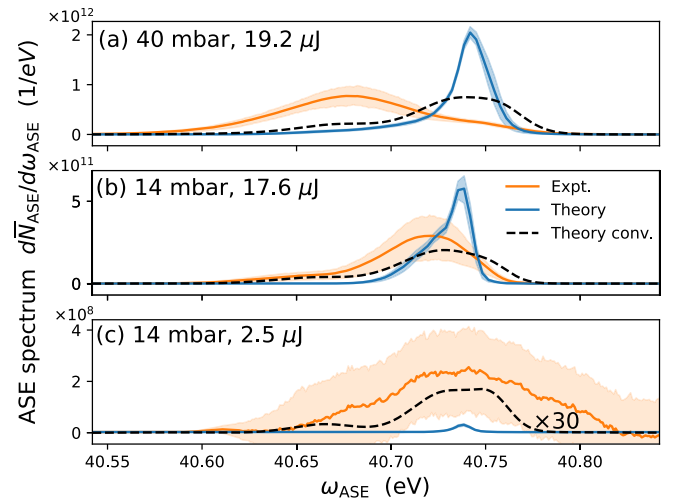


FIG. 4. The experimental and theoretical $3a \ ^1P^o \rightarrow 3 \ ^1S$ ASE spectra at three different $(p, \bar{I}_0)$ settings. Shaded areas contain 50% of the accepted pump pulses. The theory is convoluted by the WEST response function.

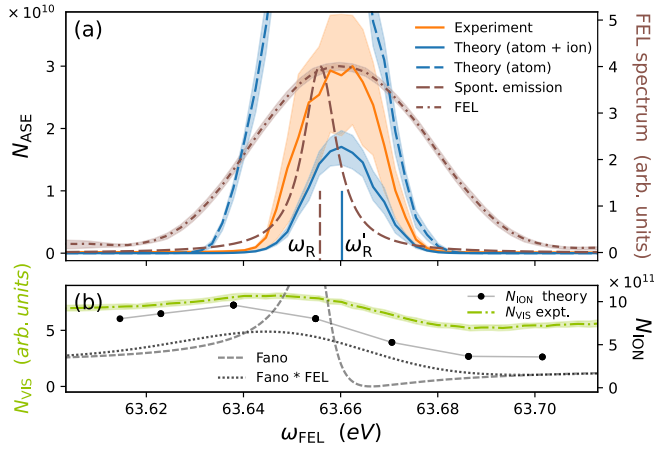


FIG. 5. (a) ASE yield as a function of the central pump photon energy ω_{FEL} in the $3a\ ^1P^o$ resonance region at $p = 15$ mbar and $\bar{I}_0 = 19.2\ \mu\text{J}$. Shaded areas contain 50% of the accepted pulses. The theoretical ASE yield (atom + ion) peaks at ω'_R photon energy. The theory version (atom), which excludes ion transition driving peaks at ω_R , the same as a Lorentzian describing spontaneous emission yield from the resonance. The pump spectrum centered at ω_R is also presented. (b) The total VIS yield N_{VIS} emitted by He gas in the cell center towards the NEWTON spectrometer mimics N_{ION} , the calculated trend of He^+ ions generated by the pump. The latter is reproduced also by the convolution of the $3a\ ^1P^o$ Fano profile [39] with the pump spectrum.

the $3a\ ^1P^o$ resonance (8 meV). The calculation reproduces the width of the ASE peak quite well (16 meV) and predicts the maximum yield at ω'_R , at a central pump photon energy slightly higher than ω_R , the $3a\ ^1P^o$ resonance energy. This frequency shift is explained by the presence of He^+ ions in the gas that may quench the ASE process. Namely, the calculated ASE field, generated by the $\text{DES} \rightarrow \text{SES}$ lasing transition in the atom (40.74 eV), may also drive the nearby $1s \rightarrow 2p$ transition in He^+ (40.81 eV). With this driving included, the model results in a five times lower ASE yield with respect to the no driving version (see Ref. [47]) and gives much better agreement with the measured ω_{FEL} dependence of ASE yield in Fig. 5(a). On the other hand, the observed dependence of the total VIS yield shows an asymmetry similar to the calculated FEL-broadened Fano profile of the $3a\ ^1P^o$ resonance [Fig. 5(b)]. The VIS emission from the gas is, therefore, dictated by the total number of He^+ ions and photoelectrons generated by the FEL pulse ionization rather than by the ASE process. This is consistent with the observed stability of the VIS spectral shape at different pump photon energies and by the presence of many He (and He^+) lines emitted in the gas cooling period that are not present in the spectrum of an isolated atom [51].

In conclusion, we have observed and quantified ASE at 30.4 nm that originates from He atoms in the short-lived autoionizing state $3a\ ^1P^o$. The inverted population was

generated by an intense resonant XUV light pulse passing through a millimeters long column of He gas at tens of millibars pressure. The theory reproduces rather well the ASE yield with regard to the pump energy and wavelength, He gas pressure, ASE spectral shape, and photon number distribution, especially at high pump energies where up to a few percent conversion to ASE was detected. Significant discrepancies were found at high gas pressure where the observed shift of the ASE spectrum is attributed to the presence of transient molecular configurations of He atoms.

Acknowledgments—We are grateful to the FERMI staff for their operation of the facility and acknowledge R. Gobessi and A. Hervat for their support in setup preparation and expert technical assistance. This work was financially supported by the Slovenian Research Agency in the framework of the P1-0112 research program and by the Joint CNR-JSI Bilateral program (2024-2025) project “Collective XUV emission from a dense gas: a route towards novel coherent light sources” (CUP: B43C24000220005). The research leading to this result has been supported by the project Laserlab-Europe under Grant Agreement No. 871124 from the EU Framework Programme for Research and Innovation HORIZON 2020. The authors acknowledge the HPC RIVR consortium and SLING for providing the computing resources of the HPC systems Arnes, NSC at JSI, and Trdina at the Faculty of Information Studies at the University of Novo Mesto.

Data availability—The data that support the findings of this Letter are openly available [52].

- [1] M. Gross and S. Haroche, *Phys. Rep.* **93**, 301 (1982).
- [2] S. Suckewer and P. Jaeglé, *Laser Phys. Lett.* **6**, 411 (2009).
- [3] W. Ackermann *et al.*, *Nat. Photonics* **1**, 336 (2007).
- [4] T. Shintake *et al.*, *Nat. Photonics* **2**, 555 (2008).
- [5] P. Emma *et al.*, *Nat. Photonics* **4**, 641 (2010).
- [6] E. Allaria *et al.*, *Nat. Photonics* **6**, 699 (2012).
- [7] N. Rohringer *et al.*, *Nature (London)* **481**, 488 (2012).
- [8] H. Yoneda *et al.*, *Nature (London)* **524**, 446 (2015).
- [9] T. Kroll *et al.*, *Phys. Rev. Lett.* **120**, 133203 (2018).
- [10] T. Kroll *et al.*, *Phys. Rev. Lett.* **125**, 037404 (2020).
- [11] M. D. Doyle *et al.*, *Optica* **10**, 513 (2023).
- [12] C. Weninger, M. Purvis, D. Ryan, R. A. London, J. D. Bozek, C. Bostedt, A. Graf, G. Brown, J. J. Rocca, and N. Rohringer, *Phys. Rev. Lett.* **111**, 233902 (2013).
- [13] O. Alexander *et al.*, *Sci. Adv.* **10**, eadp0841 (2024).
- [14] U. Bergmann, *Photosynth. Res.* **162**, 371 (2024).
- [15] A. Halavanau, A. Benediktovitch, A. A. Lutman, D. DePonte, D. Cocco, N. Rohringer, U. Bergmann, and C. Pellegrini, *Proc. Natl. Acad. Sci. U.S.A.* **117**, 15511 (2020).
- [16] R. Margraf *et al.*, *Nat. Photonics* **17**, 878 (2023).
- [17] C. Pellegrini, A. Halavanau, A. Benediktovitch, and U. Bergmann, *arXiv:2310.04019*.
- [18] K. Nakajima *et al.*, *J. Phys. Soc. Jpn.* **84**, 054301 (2015).

- [19] S. Krušič, K. Bučar, A. Mihelič, and M. Žitnik, *Phys. Rev. A* **98**, 013416 (2018).
- [20] J. R. Harries *et al.*, *Phys. Rev. Lett.* **121**, 263201 (2018).
- [21] L. Mercadier *et al.*, *Phys. Rev. Lett.* **123**, 023201 (2019).
- [22] A. Benediktovitch *et al.*, *Phys. Rev. A* **101**, 063412 (2020).
- [23] D. J. Higley *et al.*, *Commun. Phys.* **5**, 83 (2022).
- [24] L. Mercadier *et al.*, *Nat. Phys.* **20**, 1564 (2024).
- [25] S. W. J. Scully *et al.*, *J. Phys. Conf. Ser.* **58**, 387 (2007).
- [26] M. Agåker and J.-E. Rubensson, *Phys. Rev. B* **75**, 045112 (2007).
- [27] M. A. Klosner and W. T. Silfvast, *Appl. Opt.* **39**, 3678 (2000).
- [28] J.-E. Rubensson, C. Sâthe, S. Cramm, B. Kessler, S. Stranges, R. Richter, M. Alagia, and M. Coreno, *Phys. Rev. Lett.* **83**, 947 (1999).
- [29] M. K. Odling-Smee, E. Sokell, P. Hammond, and M. A. MacDonald, *Phys. Rev. Lett.* **84**, 2598 (2000).
- [30] F. Penent, P. Lablanquie, R. I. Hall, M. Žitnik, K. Bučar, S. Stranges, R. Richter, M. Alagia, P. Hammond, and J. G. Lambourne, *Phys. Rev. Lett.* **86**, 2758 (2001).
- [31] M. Žitnik, K. Bučar, M. Štuhec, F. Penent, R. I. Hall, and P. Lablanquie, *Phys. Rev. A* **65**, 032520 (2002).
- [32] L. Lipsky, R. Anania, and M. Conneely, *At. Data Nucl. Data Tables* **20**, 127 (1977).
- [33] M. Domke, G. Remmers, and G. Kaindl, *Phys. Rev. Lett.* **69**, 1171 (1992).
- [34] R. P. Madden and K. Codling, *Phys. Rev. Lett.* **10**, 516 (1963).
- [35] Š. Krušič, A. Mihelič, K. Bučar, A. Benediktovitch, S. Chuchurka, and M. Žitnik, *Phys. Rev. A* **107**, 013113 (2023).
- [36] S. Chuchurka, A. Benediktovitch, K. Špela, A. Halavanau, and N. Rohringer, *Phys. Rev. A* **109**, 033725 (2024).
- [37] Š. Krušič, K. Bučar, A. Mihelič, and M. Žitnik, *Phys. Rev. A* **98**, 013416 (2018).
- [38] Š. Krušič, K. Bučar, A. Mihelič, and M. Žitnik, *Phys. Rev. A* **99**, 013429 (2019).
- [39] A. Mihelič, Fluorescence of doubly excited states of helium in homogeneous electric field, Ph.D. thesis, University of Ljubljana, Faculty of Mathematics and Physics, Department of Physics, 2006, <http://www.rcp.ijs.si/amihelic/phd/thesis.pdf>.
- [40] J. Söderström *et al.*, *Phys. Rev. A* **77**, 012513 (2008).
- [41] L. Rumiz, D. Cocco, C. Fava, S. Gerusina, R. Gobessi, E. Mazzucco, and F. Zudini, in *Proceedings of the 2nd International Particle Accelerator Conference (IPAC 11)* (JACoW Publishing, Geneva, 2011), p. 1530.
- [42] C. Svetina, D. Cocco, N. Mahne, L. Raimondi, E. Ferrari, and M. Zangrando, *J. Synchrotron Radiat.* **23**, 35 (2016).
- [43] E. Principi *et al.*, *Nucl. Instrum. Methods Phys. Res., Sect. A* **1058**, 168856 (2024).
- [44] R. Martínez Vázquez, A. G. Ciriolo, G. Crippa, V. Tosa, F. Sala, M. Devetta, C. Vozzi, S. Stagira, and R. Osellame, *Int. J. Appl. Glass Sci.* **13**, 162 (2022).
- [45] Comsol multiphysics® v.6.2.
- [46] B. Henke, E. Gullikson, and J. Davis, *At. Data Nucl. Data Tables* **54**, 181 (1993).
- [47] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/mg5s-xvct> for spectrum background subtraction, experimental data filtering, normalization of the absolute ASE yield, normality distribution testing, and the difference between the (atom) and (atom + ion) simulations.
- [48] H. W. Lilliefors, *J. Am. Stat. Assoc.* **62**, 399 (1967).
- [49] T. W. Anderson and D. A. Darling, *Ann. Math. Stat.* **23**, 193 (1952).
- [50] S. S. Shapiro and M. B. Wilk, *Biometrika* **52**, 591 (1965).
- [51] M. Coreno, K. C. Prince, R. Richter, M. de Simone, K. Bučar, and M. Žitnik, *Phys. Rev. A* **72**, 052512 (2005).
- [52] J. Turnšek *et al.*, Data repository at Zenodo, [10.5281/zenodo.16419504](https://zenodo.org/record/16419504).