Amplification of spontaneous emission from doubly excited He atom

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Overview

Synopsis We have observed self-amplification of XUV spontaneous emission (ASE) from He atoms in 3a $^{1}P^{o}$ doubly excited state.

The 3a $^{1}P^{o}$ resonance in He with 63.66 eV

excitation energy autoionizes within 80 fs but may also decay by spontaneously emitting a 40.75 eV (30.4 nm) photon to populate the 1s3s $^{1}S^{e}$ atomic state with $3 \cdot 10^{-4}$ probability [1, 2]. Despite such a small fluorescence branching ratio, our recent calculations in the paraxial approximation predicted strong ASE in the forward direction if dense and long enough column of He gas is traversed by an intense resonant XUV pump pulse [3]. Indeed, we have observed amplification of the weak $3a \, ^1P^0 \rightarrow 1s3s \, ^1S^e$ fluorescence decay at the EIS-TIMEX beamline using light pulses from the free electron laser (FEL-1) facility FERMI in Trieste, Italy.

Images and spectra of ASE were collected, as well as secondary visible flourescence emitted in the perpendicular direction to the FEL propagation direction.

Theory background

The basic concept of ASE can be presented quasi-clasically. Let us have a gas of two level atoms, where n_u and n_l are the densities of atoms in upper and lower states. When resonant light passes through this gas, its intensity I will be governed by

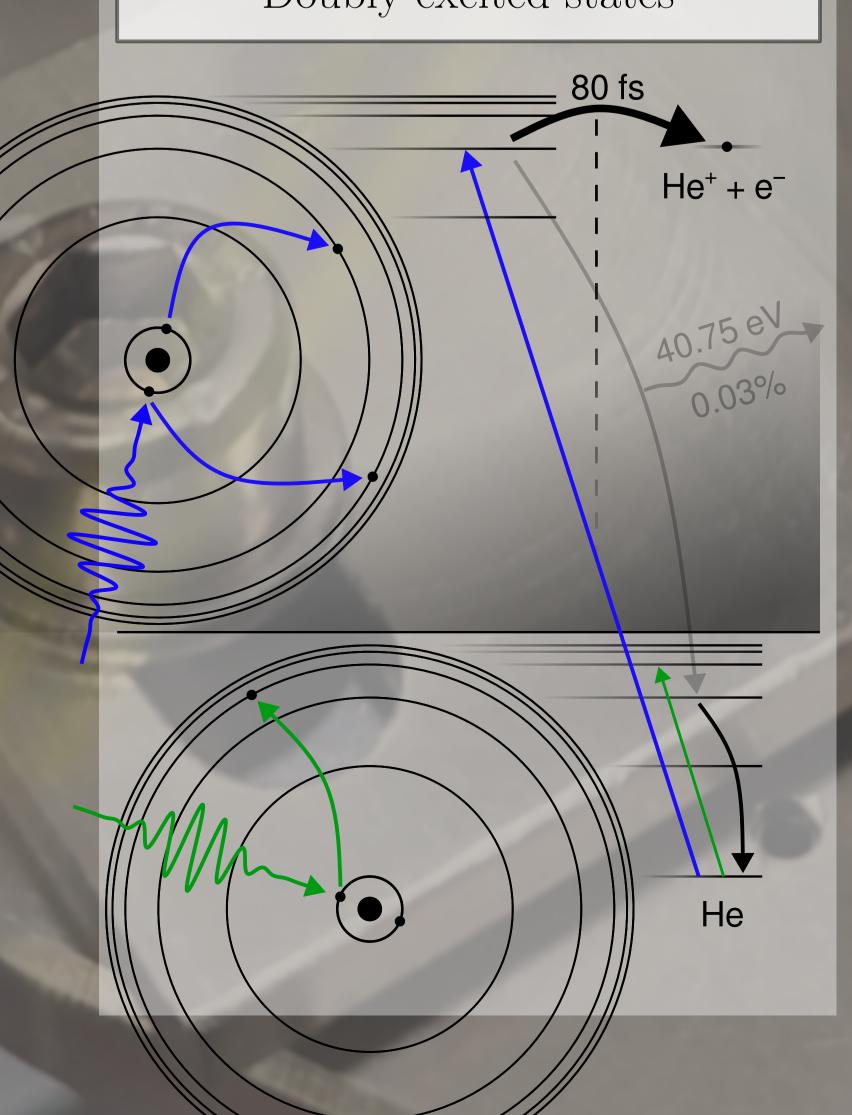
$$\frac{dI}{dz} \propto (n_f - n_i)\mu I(z) ,$$

where z is the distance along the beam and μ is the dipole matrix moment. The solution to this equation is

$$I \propto I_0 \mathrm{e}^{(n_f - n_i)\mu l}$$
.

where l is the total gas cloud length. A gas cloud prepared in a state with $n_f > n_i$ (population inversion) will thus amplify resonant light exponentially, until saturation is reached. Simulations employed a set of modified Maxwell-Bloch equations with additional stohastic noise terms in order to correctly reproduce spontaneous emission [3, 4].

Doubly excited states



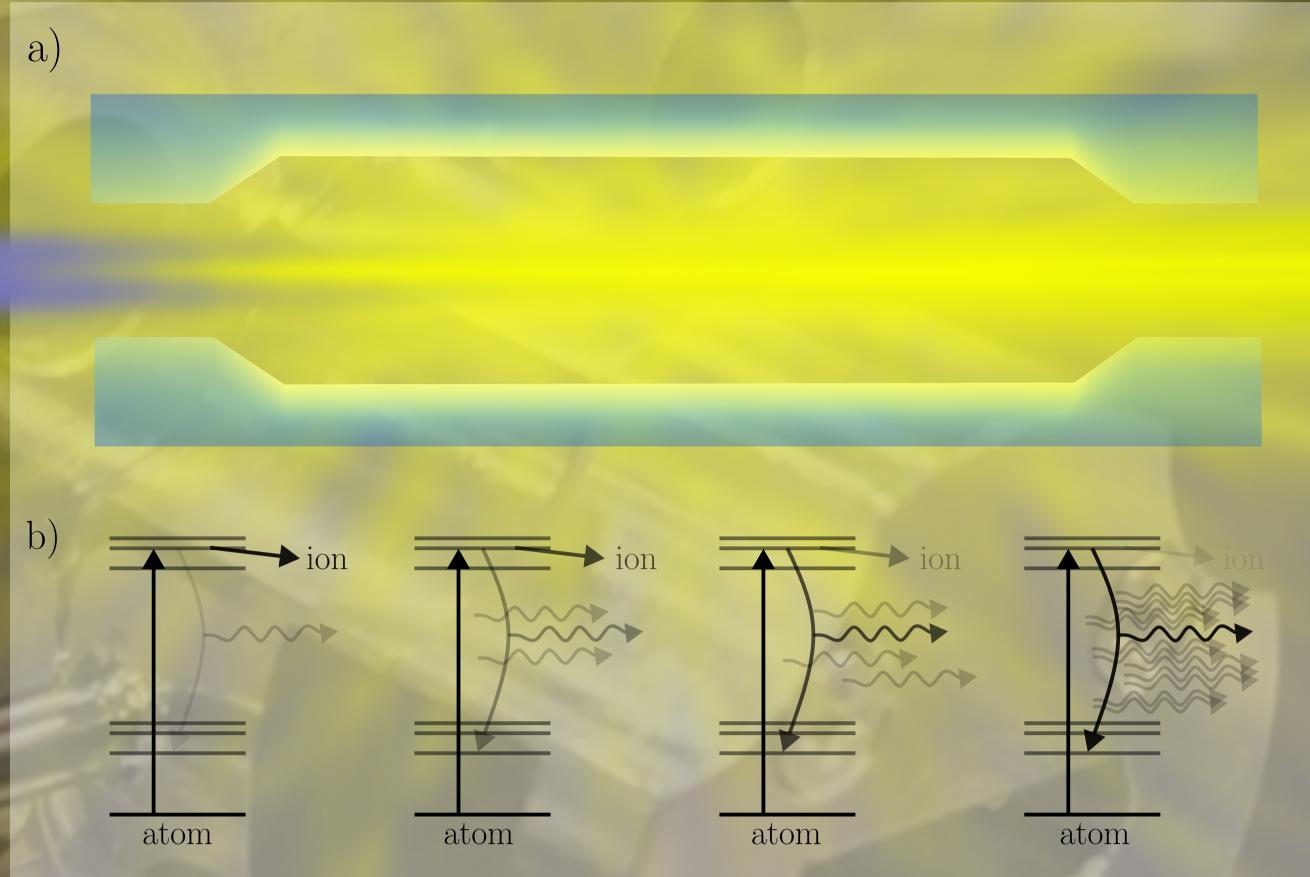


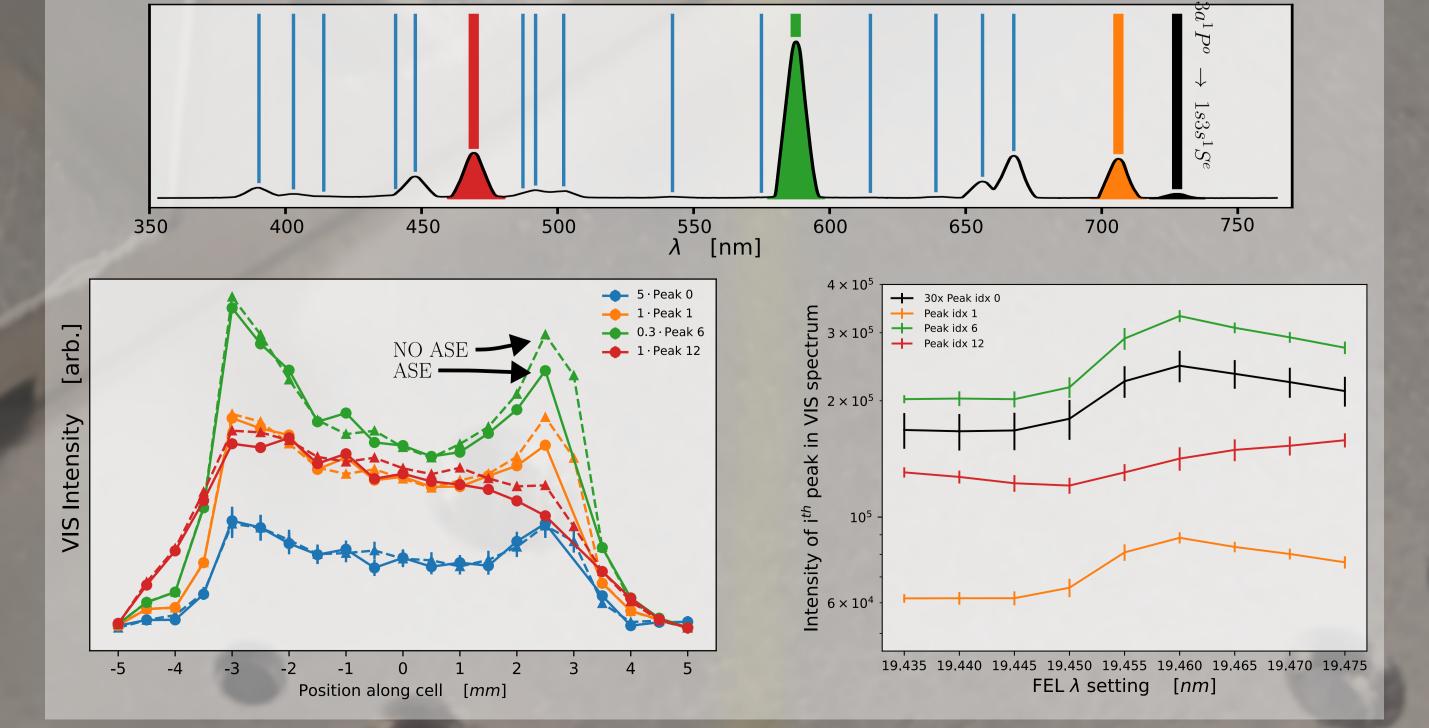
Figure 1: a) Schematic representation of the glass cell. b) S The process of stimulated emission at different positions along the cell.

Results $N_{\rm FEL}/10^{12}$ 0.50 0.75 1.00 1.25 1.50 1.75 2.00 $\times 10^{10}$ (a) Theory (atom + ion) — — Theory (atom) – Spont. emission p = 40 mbar $3a \, ^1P^o \rightarrow 3 \, ^1S$ — Expt. 10⁷ — Theory 5.0 7.5 10.0 12.5 15.0 17.5 20.0 I_0 (μJ) ····· Fano * FEL 100 - (b) 63.62 ω_{FEL} (eV) - (a) 40mbar, 19.2 μ J \bar{I}_0 (μJ) N_{ASE} (theory) (b) 14 mbar, 17 6 μ J — Theory Theory --- Theory conv. Expt. 4 (c) 14 mbar, 2.5 μ J 40.65 40.60

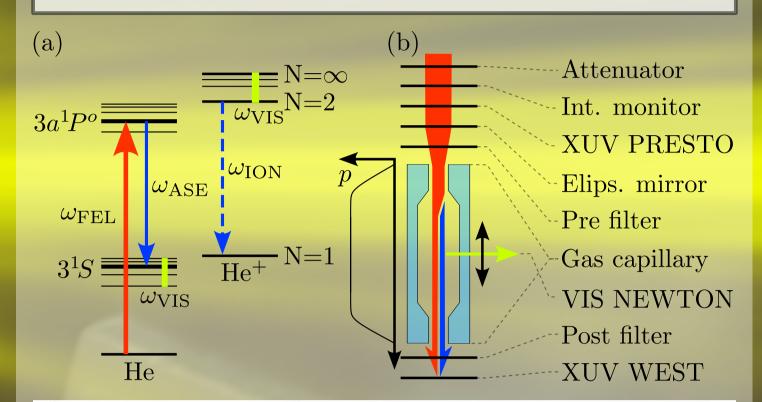
The results for the ASE part of the experiment are presented above. While the results for the VIS part of the experiment are presented below. Best conditions for ASE in this system were found to be at 40 mbar of He pressure and 20 μJ of pulse intensity. There, an average of $(4.1 \pm 1)\%$ percent of all FEL photons were converted to ASE, with the maximum single shot conversion recorded at $(6.5 \pm 1.5)\%$. Some small imprint of ASE could also be detected in the visible spectra.

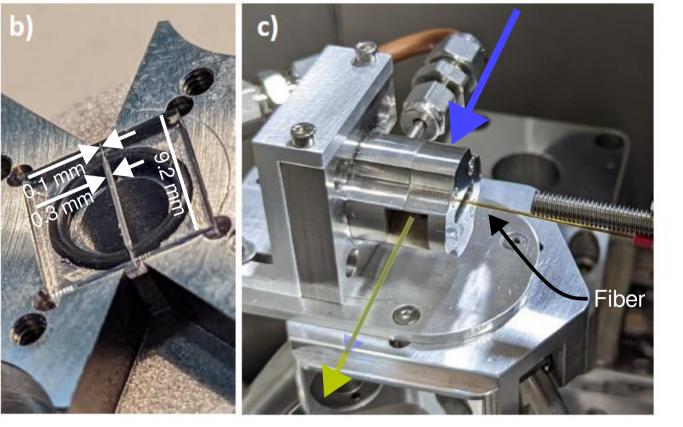
 ω_{ASE} (eV)

10



Experimental setup





Pumping of the $3a^{-1}P^{o}$ state was achieved with seeded Fermi FEL. The 50 fs long FEL pulses tuned to 63.66 eV with few tens of μJ energy were focused to the 15 \times 26 μm cross section in the center of the gas target. Incoming light was first conditioned by several thin metal filters and a gas attenuator. It was characterized by an XUV spectrometer. After the cell, the emitted spectra were recorded by another XUV spectrometer. Using an optical fiber mounted ortogonal to the FEL beam direction, we observed the visible flourescence after each pulse.

The gas target consisted of an open ended glass capillary. The glass cell consisted of a 7.2 mm long central section with 300 μm internal diameter and two 1 mm long 100 μm diameter exit holes. In this way, we achieved up to 115 mbar of helium pressure in the cell.

Outlook

A model of plasma cooling is currently in developement. It will be used to explain the measured visible fluorescence spectra in the orthogonal direction.

Upcoming experiment aims to probe other resonances for ASE emission. Also proposed is the use of resonant laser coupling between resonances, to effectively transfer populations from one resonance to the other. This would allow triggering ASE from doubly excited states that are not directly accessible from the ground state, and which could offer much more favourable fluorescence branching ratios.

Preliminary data analysis from a recent experiment might indicate, that the Fano parameter for the pumping transition could be changing due to high pressure. This presents a possible upgrade to the current theoretical model.

Bibliography

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