# First observation of 'superfluorescence' at extreme ultra-violet wavelengths

'Superradiance' was first introduced in 1954 [1] as arising from quantum correlations between excited atoms interacting through an electromagnetic field of wavelength λ, all located within a spatial region of dimension λ. Superradiance as initially described is difficult to realize in practice, but spontaneous emission can induce a related process in initially uncorrelated atoms, even in an extended medium (interatomic spacing comparable to λ, but excited atoms extending over a region in space much larger). This process was termed 'superfluorescence' [2]. Superfluorescence was first observed on rotational transitions in HF at microwave wavelengths [3], and subsequently at visible wavelengths following the availability of lasers [review in 4]. The initial stages of superradiance have been observed at X-ray wavelengths [5], and in diamond NV-centers [6].

In stark contrast to spontaneous emission, the peak intensity of superradiance scales as $N^2$, where N is the number of interacting atoms. The temporal width of the emission pulse thus scales as 1/N. N can be of the order of $10^{10}$, leading to very short, intense pulsed emission. For superfluorescence, where the initial excitation is incoherent, there is a characteristic time delay for a macroscopic polarization to develop, which also scales as 1/N. Superradiant decay can completely deplete the upper level, so potentially offers a route to intense, coherent, pulsed emission with variable pulse duration and delay. However, its generation at short wavelengths presents challenges, since higher number densities are required to meet the requirement that interatomic spacing must be comparable to wavelength. Further, suitable atomic transitions are difficult to find. The lack of suitable materials for windows and optical elements at short wavelength introduces technical constraints.

We recently overcame these challenges to observe superfluorescence at vacuum ultraviolet (164 nm) and extreme ultraviolet (30.4 nm) wavelengths for the first time [7]. A suitable atomic level scheme is found in helium ions, a high density of which can be rapidly created by ionization with free-electron laser (FEL) pulses (SACLA **BL1**) at photon energies greater than the helium ionization potential of ~25 eV (Fig. 1). Since windows cannot be used, we used a pulsed valve to inject neutral helium into a small differentially-pumped gas cell with 1 mm apertures. Essentially all of the neutral helium atoms along the FEL beam's path through the gas cell could be ionized. At 24.3 nm, resonant with excitation of the 4*p* electronic state of

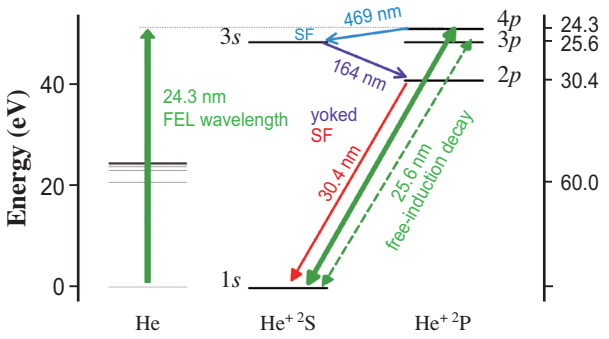


Fig.1. Partial level scheme. Helium atoms are ionized and then excited by the same FEL pulse, resonant with the 1*s*-4*p* transition in $He^+$. Due to the high density of excited 4*p* ions, superfluorescence occurs on the route 4*p*-3*s*-2*p*-1*s*, with the 2nd and 3rd steps 'yoked' due to the initial coherence of the 1*s*-4*p* excitation.

the ion, a high density of excited ions is created within the <100 fs pulse. This results in superfluorescent decay on the 4*p*-3*s* transition at 469 nm. Experimentally this was observed as highly-directional blue flashes of light (Fig. 2). Using a fast photodiode, it was confirmed that the pulses had temporal widths of the order of picoseconds, and picosecond delays with respect to the incident FEL pulse. This unequivocally confirms superfluorescence on this transition. Atomic population is transferred to the 3*s* state, which can undergo further superfluorescence decay (164 nm) to the 2*p* state. Observations with a grazing-incidence spectrometer (Fig. 2) confirmed highly-directional emission at this wavelength. Since superfluorescence requires a population inversion to proceed, further superfluorescence on the 2*p*-1*s* transition at 30.4 nm would not be expected. However, we also observed highly-directional emission at this wavelength (Fig. 2). Superradiant emission is only possible on this transition as *yoked* superfluorescence [8], which can occur if the initial excitation imparts sufficient coherence to the atomic system. Here, superfluorescence occurs from the 3*s* state to the 1*s* state 'through' the 2*p* state, with emission at 164 nm and 30.4 nm appearing simultaneously. While we were unable to confirm this interpretation experimentally due to the lack of detectors with both sufficient sensitivity and sufficient time resolution in the required wavelength regions, the intensity and directionality of the observed emissions strongly support our conclusions.

Further evidence is provided from semi-classical numerical simulations. We used the Maxwell-Liouville equations [9], with 16 atomic levels and time and spatial step-sizes of 6.5 as and 3.0 nm to model the propagation of 70-fs-long pulses through 50 μm of helium ions. The simulations were performed in parallel on the JAEA supercomputer. Results for a single pulse are shown in Fig. 3, where the rolling Fourier transform of the electric field at the output of the medium is plotted as a function of time. Free-induction decay is seen as a spectrally narrow tail at the 1*s*-4*p* resonant frequency. Similar emission is also seen at a wavelength of 25.6 nm, on the 1*s*-3*p* transition. Following a short delay (~3 ps), emission is seen at a wavelength of 469 nm, which can be interpreted as superfluorescence on the 4*p*-3*s* transition. Following a further delay (at t ~10 ps), emission is seen simultaneously at wavelengths of 164 nm (3*s*-2*p*) and 30.4 nm (2*p*-1*s*). This is consistent with our interpretation of yoked superfluorescence. Weaker signal can also be seen at wavelengths corresponding to four-wave mixing between the various wavelengths present in the medium, although these were not observed experimentally.

The observation of yoked superfluorescence is of particular interest since it arises due to the coherence of the exciting pulse, which for SACLA BL1 is only partial. From the point of view of applying coherent processes at short wavelengths, this is an important result. Whereas seeding technology (such as that used at FERMI) can be used to generate coherent FEL pulses at the wavelengths used here, shorter wavelength FELs still rely on the SASE process.

The extension to even shorter wavelengths presents the challenge of finding suitable level schemes and generating even higher target densities. One approach is to use nanoparticles such as clusters or superfluid liquid helium droplets, which offer number densities similar to the solid phase.

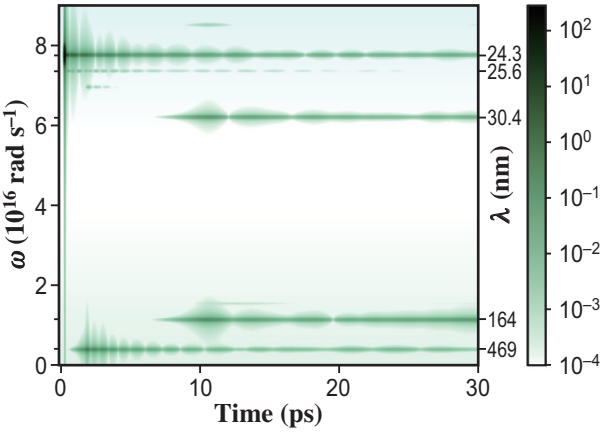


Fig. 3. Simulation results. Rolling Fourier transform (20 fs window) of the electric field at the output of the medium.

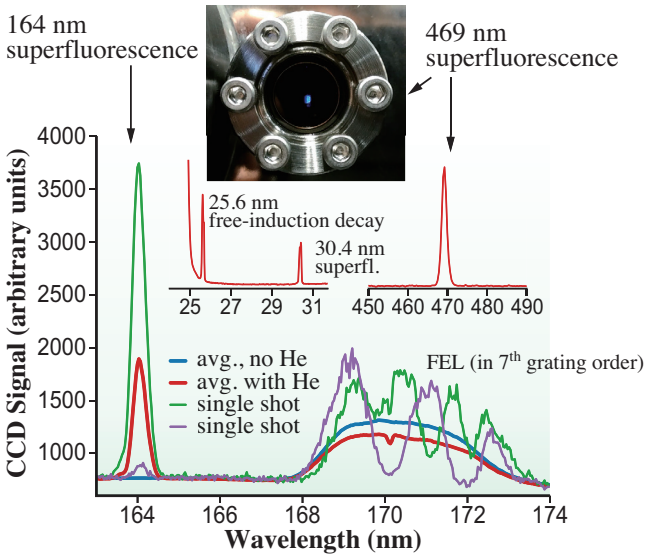


Fig.2. Overview of experimental results. Strong, highly-directional emission was observed at wavelengths of 469 nm, 164 nm, 30.4 nm, and 25.6 nm.

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