

Observation of free-electron-laser-induced collective spontaneous emission (superfluorescence)

Superfluorescence is a phenomenon in which N atoms in excited states (N is a number greater than one) decay collectively and simultaneously to produce a burst of highly-directional radiation with a peak intensity many times higher ($\propto N^2$) and duration many times shorter ($\propto 1/N$) than would be observed if the atoms were to decay individually. First described by Dicke in 1954 [1], the effect shows similarities with laser emission (high directionality and high coherence) but is fundamentally different since the decay is **collective**, and can only occur if 1) the spacing between the atoms is less than λ , the wavelength of the emitted radiation, and 2) the population inversion is created near-instantaneously (see below).

Since the first description of the phenomenon, superfluorescence has been observed for both very large ($N > 10^{12}$) and very small ($N = 2$) collections of atoms, for emitted wavelengths ranging from a few hundred to a few thousand nanometers, and on timescales ranging from picoseconds to microseconds. The physics behind the phenomenon is of direct relevance to many effects in quantum optics [2]. To observe the phenomenon a population inversion must be created in an ensemble of atoms on a timescale shorter than the superfluorescence decay, and in this work we have used pulses of 100 fs duration from the RIKEN SCSS free-electron laser (FEL) facility located at SPring-8. The FEL radiation was focussed to a spot of diameter $\sim 10 \mu\text{m}$ at the center of a 44 mm sided cubic gas cell (see Fig. 1), into which helium gas was introduced via a pulsed nozzle synchronized to the 30 Hz FEL pulses. This setup allowed us to

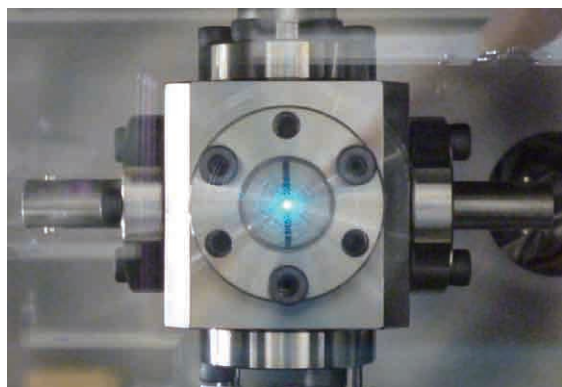


Fig. 1. Photograph of a single flash of 501.6 nm superfluorescence from helium at the centre of a gas cell, as viewed through a glass window at the end of the SCSS beamline.

maximize the number density of ground state atoms present at the time of each FEL pulse, and also control the density of these atoms by varying the gas pressure behind the nozzle. Each FEL pulse then created up to 10^8 helium atoms in $1s3p$ excited states (see Fig. 2) within a volume of less than 1 mm^3 .

Intense bursts of superfluorescence were observed as the atoms simultaneously decayed to the $1s2s$ state emitting 501.6 nm radiation. Using a calibrated photodiode we determined that up to around 2.5×10^{11} photons were produced in each superfluorescence pulse – a photon number conversion efficiency of around 10%. The emitted radiation was found to be highly directional, with an angular divergence only slightly larger than that of the FEL radiation itself.

Figure 2 shows a simplified level scheme of the neutral helium atom. Helium has two electrons, and in the ground state both electrons are in $1s$ states. The wavelength of the FEL radiation was set to 53.7 nm, which corresponds to the $1s1s$ to $1s3p$ transition. Due to the ‘spiky’ nature of the SASE pulses the overlap with the transition varied shot-by-shot. The radiative lifetime of the $1s3p$ state is about 1.7 ns, which means that if an excited population (N) of atoms in the $1s3p$ state were to decay **independently**, a histogram of decay times with respect to excitation would show an exponential decay, with the upper state population dropping from N to N/e after 1.7 ns. In the independent decay regime, an excited helium atom in the $1s3p$ state decays with 97% probability back to the $1s1s$ ground-state, with only around 3% of the decays populating the metastable $1s2s$ state and leading to fluorescence at 501.6 nm. When the inter-atomic spacing is less than 501.6 nm, however, as was the case in our experiments where the number density of neutral atoms in the gas cell was up to around 10^{17} cm^{-3} (a gas pressure of 3000 Pa), **collective** superfluorescence decay can occur on this transition. In this process the entire population of interacting atoms is transferred from the $1s3p$ state to the $1s2s$ state. The superfluorescence pulse width is given by $\tau = 8\pi / (3\lambda^2 \Gamma \rho L)$, where $\lambda = 501.6 \text{ nm}$, Γ is the natural decay rate ($1.3 \times 10^7 \text{ s}^{-1}$), ρ is the number density of excited atoms ($\approx 10^{17} \text{ cm}^{-3}$), and L the length of the extent of the inversion ($< 44 \text{ mm}$). As well as a temporal width much shorter than that for independent decay, superfluorescence is characterized by a delay between excitation and emission of the pulse. This is given by $\tau \times (\ln(2\pi N))^{1/2} / 4$, where τ is the pulse width defined above. This delay can be understood as the time required for N

oscillating dipoles to become in-phase due to the coupling through the electromagnetic field, and has a statistical variation shot-by-shot. It can be seen from the two equations that both the width and the delay scale as $1/N$. Since the width of the pulse scales as $1/N$ it is clear that the peak intensity must scale as N^2 , in order to conserve photon number.

To characterize the observed superfluorescence we used a streak camera to record the time profile of the pulses shot-by-shot. An overview of the results is given in Fig. 3. Each trace is the average of several streak camera traces recorded at several different gas pressures P , ranging from 0.5 atm to 9 atm. These gas pressures correspond to pressures in the gas cell of around 300 Pa to around 3000 Pa, and a proportional variation of N , the number of excited atoms. The general trend is that higher pressures (larger N) lead to narrower pulses with shorter delays. The inset to the figure shows that whereas the total intensity scales as P^1 (and thus N^1), the peak intensity scales as P^2 (N^2), consistent with superfluorescence. A more detailed analysis reveals that the width and delay both scale as $1/N$ [3].

This work represents the first observation of superfluorescence following excitation in the extreme ultraviolet wavelength region, and also the first observation using FEL pulses as the excitation source. Extension by suitable choice of system suggests a way of generating tailor-made pulses in the EUV and even X-ray wavelength regions which are 'cleaner' than the initial SASE-derived pump pulses, and complements more traditional atomic laser techniques [4]. A further tantalizing

prospect is that of superfluorescence occurring in large molecular systems, where for example a large number of C atoms are separated by less than the C $2p \rightarrow 1s$ transition wavelength of 4 nm.

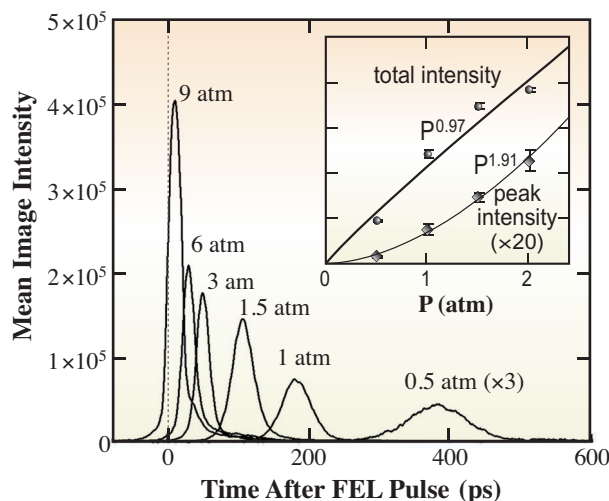


Fig. 3. Representative streak camera traces. Each trace is the average of several traces recorded at each gas pressure (proportional to the number of excited atoms N). The inset shows the pressure dependence of peak and total intensity [3].

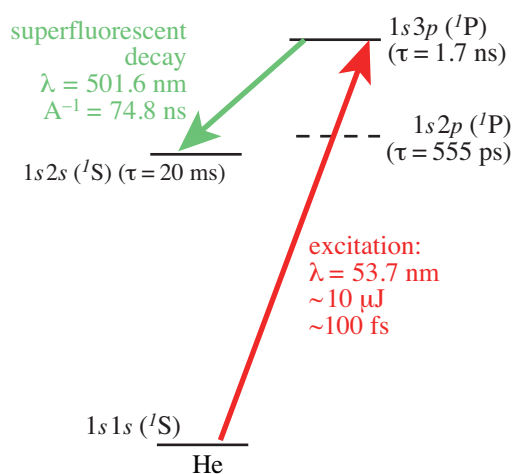


Fig. 2. Simplified level scheme of neutral helium. FEL pulses with a centre wavelength of 53.7 nm (23.1 eV) can excite atoms to the $1s 3p$ state. Superfluorescence is observed at 501.6 nm on the $1s 3p \rightarrow 1s 2s$ transition [3].

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