Chapter 16
Superfluorescence/Superradiance in Helium Following Free-Electron Laser Excitation

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Abstract We discuss the observation of superfluorescence following the excitation of helium atoms with pulses from a free-electron laser. From semi-classical simulations and consideration of transition parameters, we predict that it should be possible to generate pulses of EUV superfluorescence using two-photon excitation.

16.1 Introduction

Superradiance is a fundamental effect which can occur when ultrafast, intense radiation is used to excite dense atomic samples. Its development is sensitive to the spectral and coherence properties of the incident radiation, and as such is an excellent test of our understanding of these properties. This is particularly relevant with the increasing availability of coherent light sources operating at EUV and X-ray wavelengths. First discussed by Dicke [1], superradiance occurs following the creation of a macroscopic polarization by coherent excitation. With incoherent excitation the related process superfluorescence can occur, where the macroscopic polarization emerges spontaneously, leading to a characteristic and stochastic delay [2–4]. Superfluorescence has been observed in gaseous and solid media, Bose–Einstein condensates, and nanometre-sized structures [5], but never (to our knowledge) at wavelengths significantly shorter than the visible, although an effect related to superradiance has been observed at X-ray wavelengths [6]. Recently, we reported the observation of superfluorescence at visible wavelengths following the excitation of helium atoms using intense pulses of radiation from a free-electron laser (FEL),

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resonant to 1s3p (53.7 nm) and 1s4p (52.2 nm) excitation [7–10]. Simulations (unpublished) have also shown that the partial coherence of the FEL pulses may also lead to superradiant decay of the 1snp states back to the ground state, at the same EUV wavelengths. However, the experimental verification of this is difficult since the emitted radiation is at the same wavelength as the intense excitation pulse. Here we propose making use of the partial coherence of the excitation and using two-photon excitation to induce yoked superfluorescence [11–14], leading to the emission of superfluorescence at wavelengths in the EUV region and at shorter wavelengths than the excitation pulse.

16.2 Yoked Superfluorescence

To generate superfluorescence at EUV wavelengths we propose using two-photon excitation to excite either the 1s3s/1s3d or 1s4s/1s4d states. To demonstrate that this is feasible using pulses typical of a free-electron laser, Fig. 16.1 shows the results of a simulation of the interaction of a 4 μJ FEL pulse with a duration of around 50 fs and a central wavelength of 107.4 nm. To simulate the partially coherent nature of FEL pulses the partial coherence method of Pfeifer et al. was used [15]. For details of the simulation methods see [16, 17]. For this particular pulse, it is clear that significant excitation to 1s3d can be expected, along with some excitation of 1s3s, and also 1s4s, 1s4d. Different realizations of the stochastic pulses lead to different final populations, but at this central wavelength the majority of atoms are either excited to 1s3d or remain in the ground state. While the results are not shown here, similar simulations with an excitation wavelength of 104.4 nm suggest that the most likely state to be excited is 1s4d, with 1s4s excitation weaker, but stronger than is 1s3s excitation at 107.4 nm.

Having established that it is feasible to create high densities of helium atoms in the 1s3d or 1s4d states, we discuss whether yoked superfluorescence can be expected

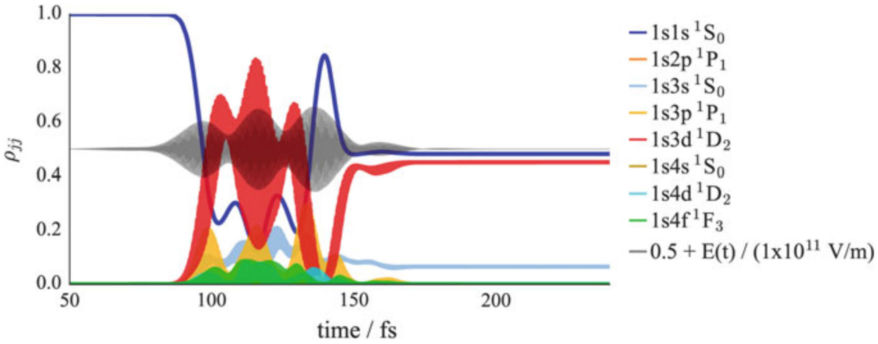

Fig. 16.1 Simulation of the two-photon excitation process with a partially coherent FEL pulse (see text), showing the evolution of the populations of selected levels. The black trace shows the electric field, centred and scaled

Table 16.1 Wavelengths, characteristic superfluorescence times and threshold column densities for the relevant transitions

Upper	Lower	λ/nm	$\sigma\tau_{SF}$/(ps nm^{-2})	σ_{th}/nm^{-2}
1s3s	1s2p	728.3	0.86	1.2
1s3d	1s2p	668.0	0.29	0.37
1s4s	1s2p	504.9	4.9	4.6
1s4s	1s3p	2114	0.41	1.6
1s4d	1s2p	492.3	1.7	1.6
1s4d	1s3p	1909	0.32	1.2
1s2p	1s1s	58.4	1.4	0.15
1s2p	1s2s	2059	1.0	3.9
1s3p	1s1s	53.7	5.1	0.52
1s3p	1s2s	501.7	2.5	2.4
1s3p	1s3s	7438	0.60	8.5
1s3p	1s3d	96000	5.9	19

to occur. Table 16.1 shows some parameters of the relevant transitions. The quantity $\sigma\tau_{\mathrm{SF}}$ is the sample column density $\sigma = \mathrm{N}_a\mathrm{L}$ multiplied by the characteristic superfluorescence time for a cylindrical sample $\tau_{SF} = 8\pi/(3\lambda^2\mathrm{A}_{ki}\mathrm{N}_a\mathrm{L})$, where N_a is the number density of excited atoms, L the length of the gain medium, λ the wavelength of the transition, and A_{ki} the spontaneous decay rate (Einstein A coefficient). The quantity $\sigma\tau_{SF}$ is thus a means of comparing the superfluorescence decay rate for different transitions given the same excited state column density. The final column, σ_{th} is the threshold column density defined by equating the superfluorescence decay rate to the wavelength-dependent loss rate due to diffraction (see [16] for details), which is the dominant loss rate for the conditions considered—a cylindrical sample of length 2 mm and diameter 12.6 µm. For yoked superfluorescence to proceed on a particular two-step decay requires [11–14] that (i) the first step transition is that most likely to proceed as superfluorescence, and (ii) the superfluorescence decay rate of the second step is not significantly faster than that of the first step. For the gain medium assumed, the threshold excited state number density required for superfluorescence to proceed on the 1s3d-1s2p transition (the only possible decay route from 1s3d) is around 2×10^{20} m^{-3}. An excitation probability of 0.5 would thus require a ground state number density of 4×10^{20} m^{-3}, well within the range of densities accessible experimentally. The threshold column density for 1s2p-1s1s is significantly lower, and the superfluoresence decay rate is longer, leading to yoked superfluorescence (excitation-induced coherence favours 1s1s over 1s2p). Transitions from the 1s4d and 1s4s states have higher thresholds, but are still well within reach experimentally, and the 1s4d/4s-1s3p-1s1s yoked superfluorescence schemes can also be expected to occur. Preliminary Maxwell–Bloch simulations considering the propagation of the field through the medium suggest that the predicted yoked superfluorescence does indeed occur, and experiments are planned at the SACLA BL1 free-electron laser light source to confirm the predictions. Assuming that the pulse energies, pulse durations, and focus sizes available at a wavelength of 107.4 nm are comparable to those used in single-photon experiments [7], the emissions following two-photon excitation can be expected to be similar in intensity to those following single-photon excitation, since in both cases it is predicted that similar excited state densities can be created, leading to the first observation of superfluoresecence at EUV wavelengths.

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