放射光科学研究センター　コヒーレントX線利用研究グループ

軟X線領域における原子のコヒーレントコントロールに向けての研究

Towards atomic coherent control with X-rays

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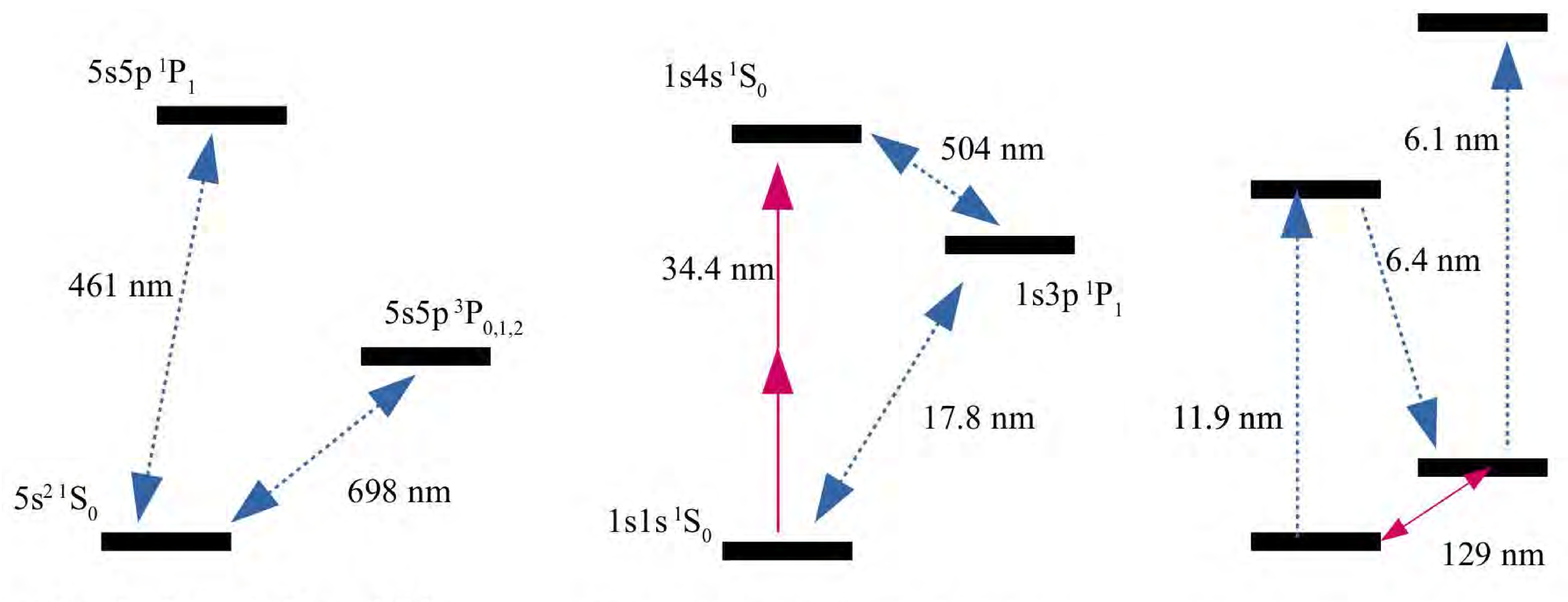
研究概要

Lasers operating close to the visible wavelength region are ubiquitous both in society and as research tools. One of the limits of their application to new technologies and research techniques is the short wavelength barrier, comprising the lack of reflective optics and the technological problems behind the generation of coherent radiation in this region.
Current laser-like sources at short wavelengths include plasma-based X-ray lasers, visible-laser based high-harmonic generation, and accelerator-based free-electron lasers. While only partially coherent, X-ray free-electron lasers offer high intensity, short pulses, and wavelength selectability.
The goal of the current research is to develop quantum optics techniques which can form the basis of new research tools, and have the potential to become new analytical techiques. For example, figure a) shows a visible-wavelength atomic scheme used for atomic clocks. Extending this to shorter wavelengths would lead to improved accuracy. Schemes such as that of c) have been proposed as new probes for molecular excitations. Recent work is aimed at extending previous research at EUV wavelengths to a similar scheme at shorter wavelengths (b).



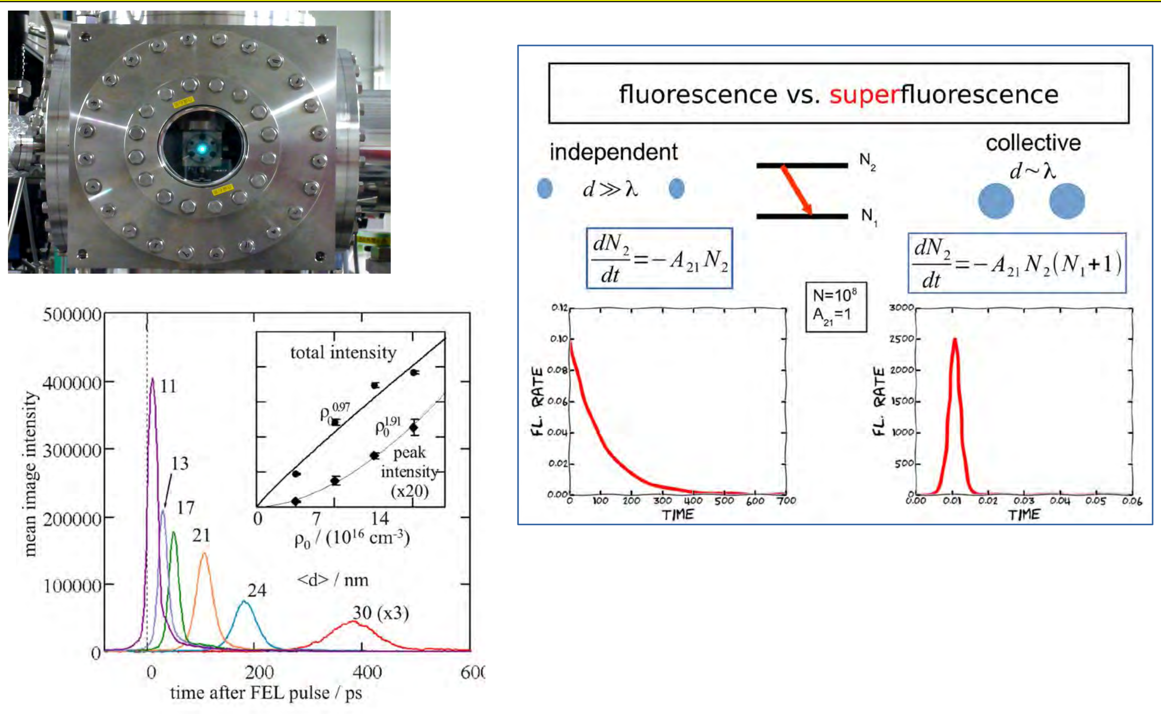
a) level scheme in Sr at visible wavelengths, used for state-of-the-art atomic lattice clocks

b) Isolated level scheme at near-x-ray wavelengths in Li+, proposed here for generating superfluorescence at 17.8 nm

c) Level scheme for X-ray 4-wave mixing (Tanaka and Mukamel)

研究成果

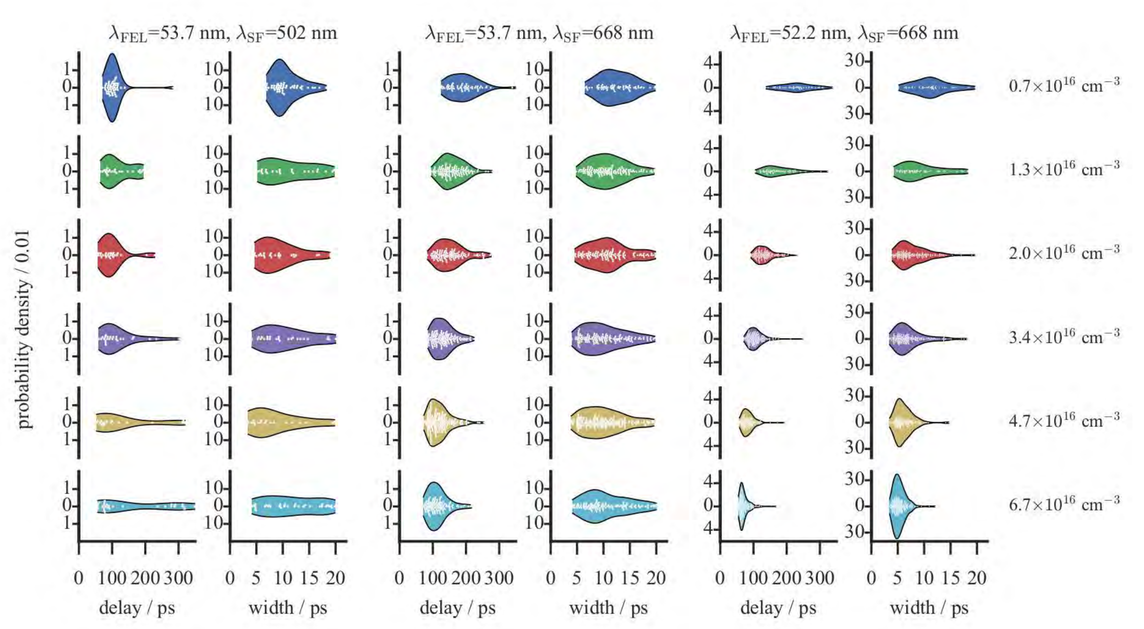
1. Superfluorescence in helium at 502 nm



Superfluorescence can occur when a population inversion is rapidly and incoherently created on an atomic transition. It is strongly related to the superradiance phenomenon (Dicke, 1954), which requires coherent excitation. Due to the high density neighbouring excited atoms can not decay independently. Constructive interference leads to a rapid depletion of the upper level in a superfluorescence burst, with a peak intensity which scales as N^2, where N is the number of excited atoms. A stochastic delay time is required for the build-up of the macroscopic dipole moment – this is characteristic of superfluorescence (in contrast to superradiance and amplified spontaneous emission), and is clearly seen in the experimental results.

Nagasono M, Harries J R et al 2011 Phys. Rev. Lett. **107** 193603

2. Superfluorescence in helium at 668 nm, 728 nm



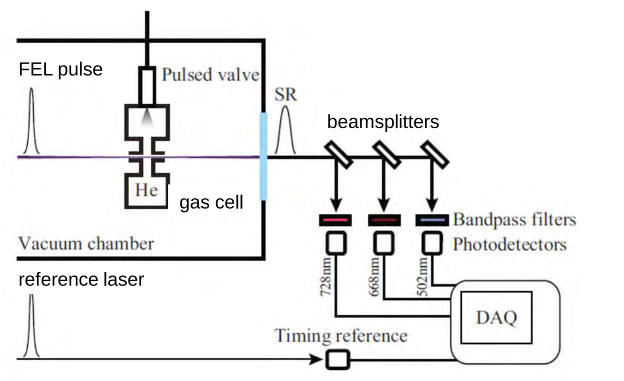


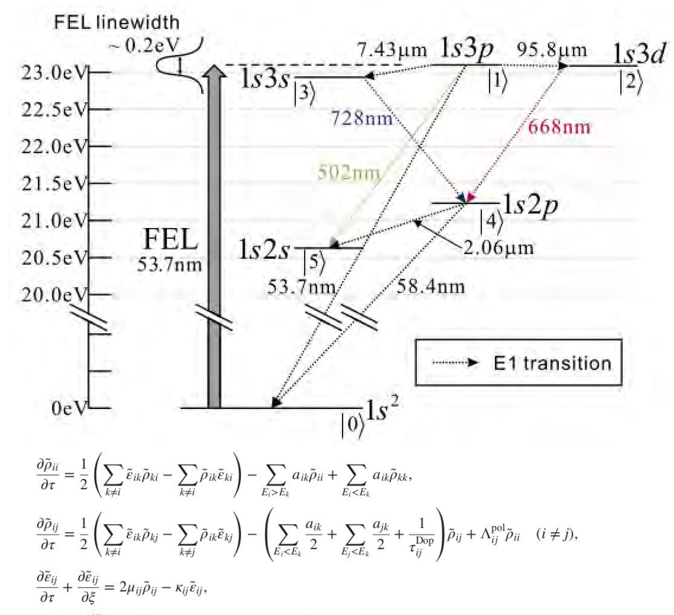
TABLE I. Wavelengths, characteristic superfluorescence times, and threshold column densities for some singlet transitions. The threshold excited atom column density is estimated by equating τ_{SF} and T_{diff} (see text) for a diameter of 12.6μm.

upper	lower	λ/nm	$\sigma\tau_{SF}/(\mathrm{ps\,nm^{-2}})$	$\sigma_{th}/\mathrm{nm^{-2}}$
1s3p	1s1s	53.7	5.13	0.52
	1s2s^a	501.7	2.49	2.4
	1s3s	7438	0.60	8.5
	1s3d	95787	5.98	19^b
1s3d	1s2p^a	668.0	0.29	0.37
1s3s	1s2p^a	728.3	0.86	1.2

^{a} transition experimentally observed as superfluorescence [4–6]
^{b} Using $T_{diff} = \lambda/c$

Nakajima K, Harries J R et al 2015 J. Phys. Soc. Jpn. **84** 54301
Harries J R et al 2015 J. Phys. B: At. Mol. Opt. Phys. **48** 105002

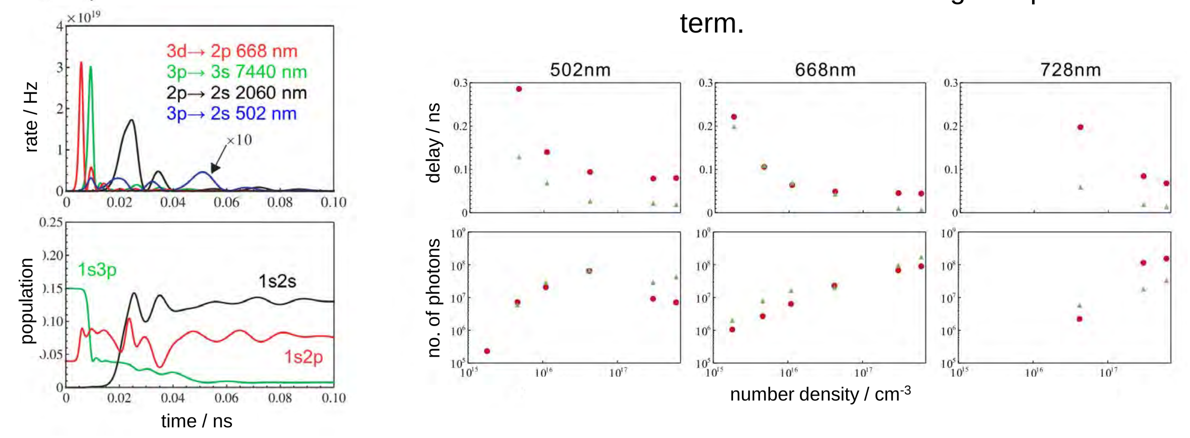
3. Maxwell-Bloch propagation simulations (i)



Assuming initial population is created in the 3s, 3p, and 3d levels, the Maxwell-Bloch equations were used in the rotating-wave and slowly-varying envelope approximations to model the development of superfluorescence.

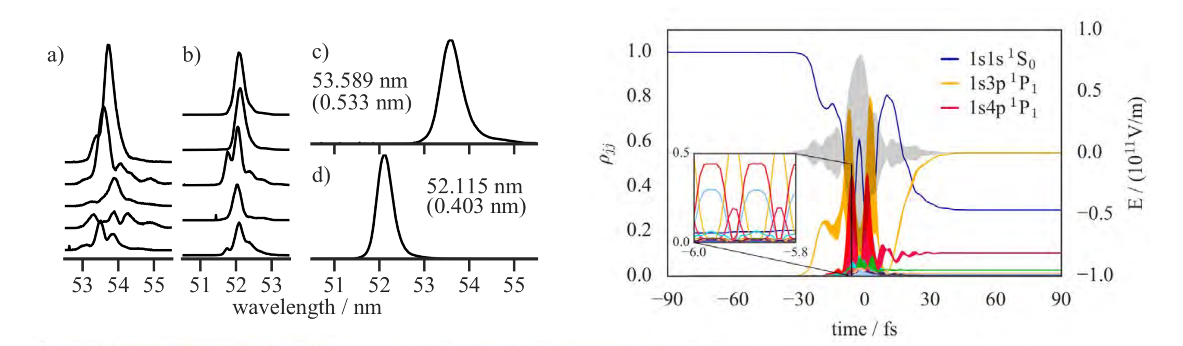
The propagation of the electric field E(t, z) was modelled through an assumed-cylindrical atomic medium. Spontaneous emission is represented by a random polarisation term – this initiates the superfluorescence with a stochastic delay.

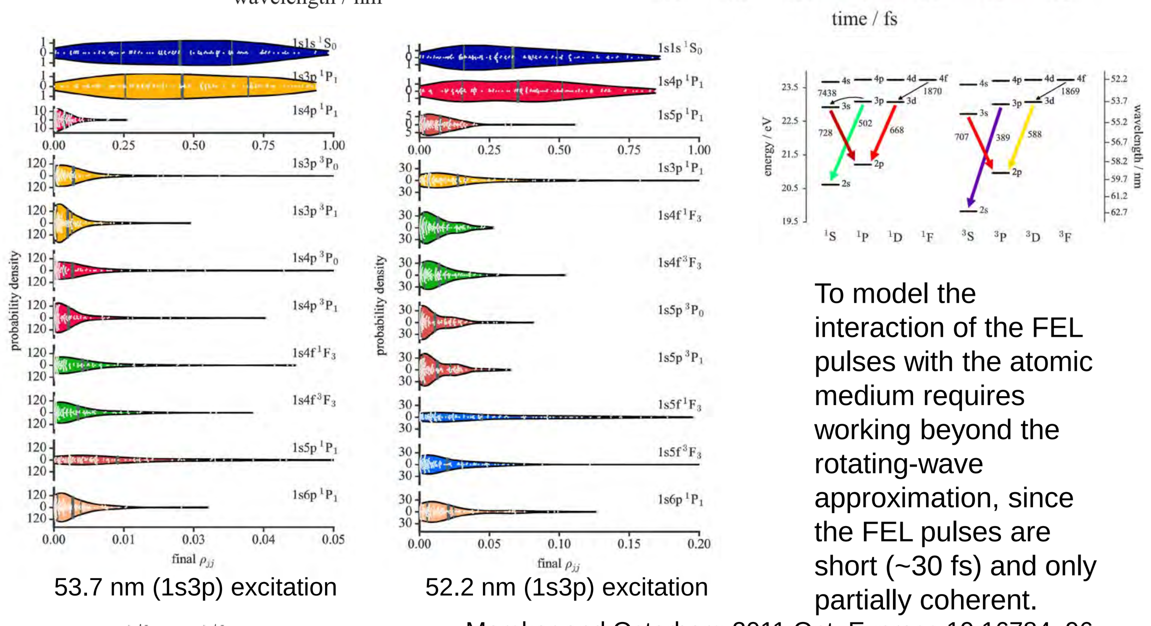
Diffraction losses are included in the model with a wavelength-dependent term.



Ohae C, Harries J R et al 2016 J. Phys. Soc. Jpn. **85** 34301

4. Beyond the rotating-wave approximation





53.7 nm (1s3p) excitation　　52.2 nm (1s3p) excitation

To model the interaction of the FEL pulses with the atomic medium requires working beyond the rotating-wave approximation, since the FEL pulses are short (~30 fs) and only partially coherent.

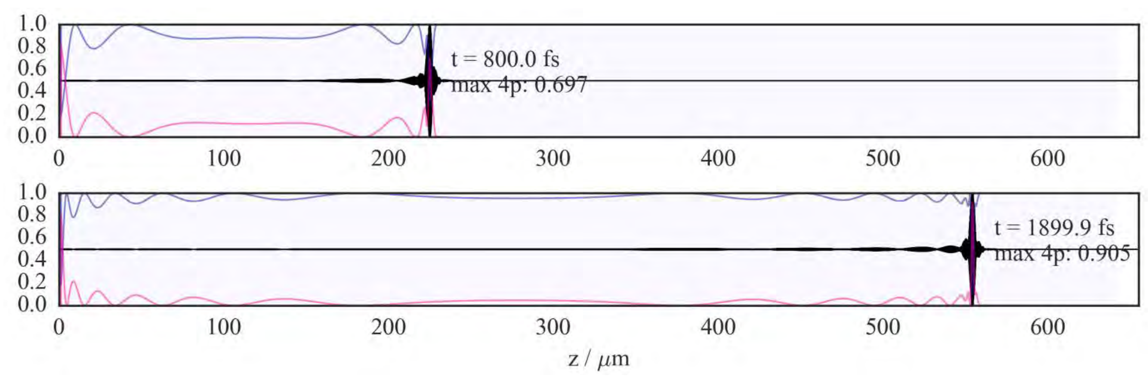
$$|\rho^{n+1}\rangle\rangle = \check{U}_0^{1/2}\check{U}_I^n\check{U}_0^{1/2}|\rho^n\rangle\rangle$$

Δt = 8 as

Marskar and Osterberg 2011 Opt. Express 19 16784–96
Pfeifer T et al 2010 Opt. Lett. 35 3441–3

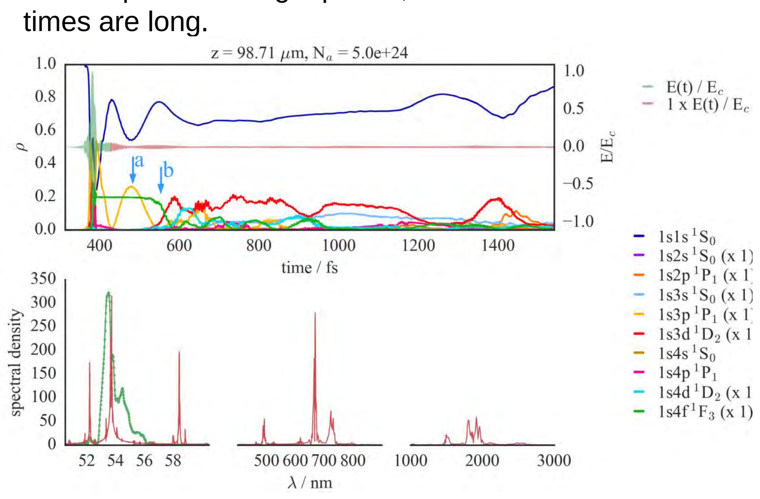
Harries J R et al 2016, submitted to Phys. Rev. A

5. Maxwell-Bloch beyond the rotating-wave



Short wavelengths (~50 nm) and long propagation distances (2 mm = 40 000 λ) make calculations challenging, particularly for multiple levels. Pseudosepctral methods reduce stepsizes to several per wavelength/period, but calculations times are long.

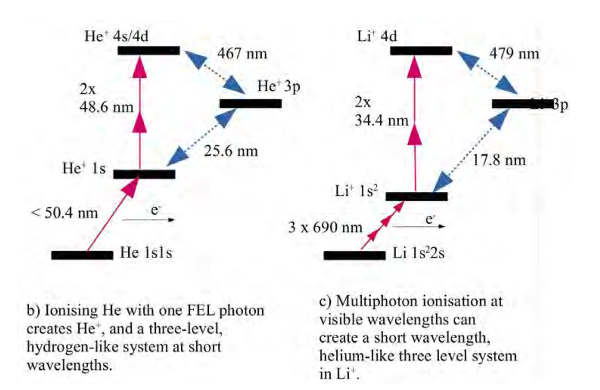
This 2-level calculation with a Gaussian input pulse of similar duration to the FEL pulses reveals an SIT effect.



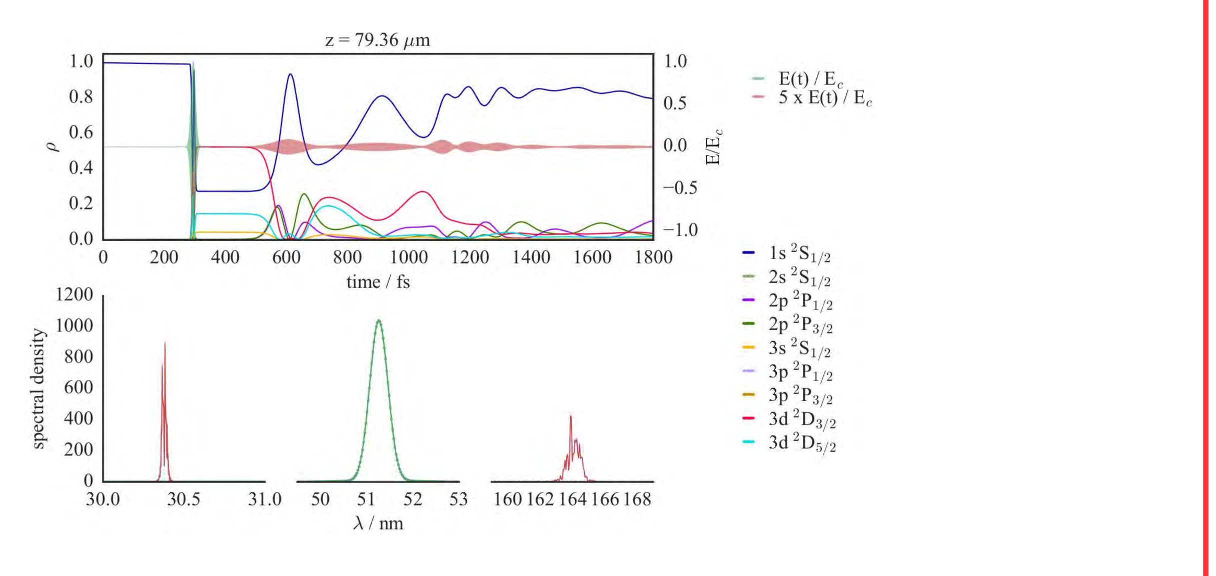
This multi-level simulation using a simulated FEL pulse reveals the development of amplified spontaneous emission and cascade superfluorescence

Harries J R et al 2016, in preparation

6. Prospects for multi-photon excitation at SACLA



Coherent two-photon excitation can lead to *yoked* superfluorescence, where two cascade decays are inter-dependent. This is a potential route to creating superfluorescence at shorter wavelengths than the incident radiation.



Harries J R et al 2016, in preparation

まとめ

Coherent lightsources at short wavelengths are currently being developed at rapid rates. To bridge the gap between technical advance and real-world applications requires the understanding of the interaction between these new lightsources and matter, beginning from the most fundamental levels of the electric field interacting with atomic levels.

Using the phenomenon of superfluorescece/superradiance as an example, we have successfully shown that a) pulses from current SASE (self-amplification of spontaneous emission) free-electron laser sources are short enough and intense enough to observe fundamental quantum optics effects, and b) that an understanding requires many-level treatments going beyond the rotating-wave and slowy-varying envelope approximations.

New experimental and calculational techniques have been developed to continue this research.

今後の課題

- Combination of X-ray streak camera and X-ray spectrometer at SACLA BL1
- many-level simulations of short-wavelength pulses require parallel computer time
- multi-photon excitation schemes will use 2-pulse operation, or externally triggered laser
- Demonstration of quantum optical effects leading to applications will require synchronised, intense, femtosecond and nanosecond tunable lasers operating at visible wavelengths and shorter

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