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研究課題名（和文）Development of a dense source of highly-charged ions for X-ray quantum optics

研究課題名（英文）Development of a dense source of highly-charged ions for X-ray quantum optics

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研究成果の概要（和文）：本研究の目的は、「多価イオン」、すなわち多くの電子を失った原子、の高密度な試料を作成し、短波長コヒーレント光源（X線自由電子レーザー）を用いて、様々な量子光学現象の実現である。又、その実験結果の理解のための数値シミュレーションも目的の一つとしている。主の成果は、i)自由電子レーザー用中性、イオンの試料生成のためのガスセル開発、ii)中性ヘリウムにおける自由電子レーザー光のラマン散乱およびFID蛍光の観測、iii)水素様ヘリウム（He+）における「超蛍光」の観測、iv)それぞれの現象の数値計算シミュレーション、とv)自由電子レーザーおよび放射光利用に向けた電子ビームイオントラップの開発。

研究成果の概要（英文）：The goals of this research are to prepare samples of 'highly-charged ions' (atoms which have lost a large fraction of their electrons) to study various quantum optics effects using short-wavelength coherent lightsources (X-ray free-electron lasers). Further, the development of numerical simulations to support this experimental research was also a goal. The main results include i) development of a gas cell for creating a high density of neutral atoms and ions, ii) observation of Raman scattering and free-induction decay following excitation of neutral helium with a free-electron laser, iii) observation of 'superfluorescence' from hydrogen-like helium (He+), iv) numerical simulations of these processes, and v) further development of an electron-beam ion trap suited to studies at free-electron laser and synchrotron lightsources.

研究分野：コヒーレントX線利用研究、原子分子

キーワード：原子・分子　量子光学　超蛍光　超放射　多価イオン

１．研究開始当初の背景

The scientific goal of the main focus of this research is the extension of quantum optical effects well-established at visible wavelengths to the extreme-ultraviolet and shorter. The potential advantages of working at shorter wavelengths are numerous, including greater element and site selectivity for analysis techniques, more efficient single-photon detection, and the possibilities of greater precision for metrology using atomic/ionic lines. Whereas at visible wavelengths intense, tuneable coherent lightsources are readily available in the form of the various laser technologies, lightsources with similar properties in the EUV and soft X-ray regimes may regarded as still being in their infancy. The applicant has concentrated on the use of free-electron laser lightsources, which offer partially coherent, femtosecond, highly intense pulses of light at EUV wavelengths and shorter.

２．研究の目的

The goal of the current project was to focus on preparing the dense atomic/ionic targets necessary to observe effects such as superfluoresence at EUV wavelengths. To this end, the aim was to develop techniques of generating dense sample of neutral atoms and ions at the focus point of a free-electron laser beamline. Due to the target wavelengths, windows cannot be used, and a major consideration is maintaining the high vacuum of the beamline.

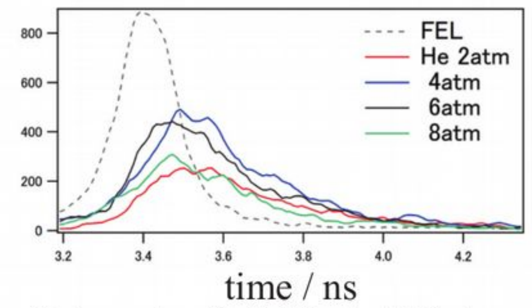
３．研究の方法

Two directions were pursued: i) using the free-electron laser pulse itself to ionise neutral atoms and create instantaneous high populations of ionic targets, and ii), developing an electron-beam ion trap collinear to the beamline. In approach i), experiments were performed at BL1 of the ‘SACLA’ X-ray free electron laser facility. Approach ii) was carried out in collaboration with a group mainly working at the Max-Planck Insitut fur Kernphysik, Heidelberg, Germany.
In parallel to the experimental approaches, numerical simulation techniques were developed to study the interaction of partially-coherent free-electron laser pulses with dense atomic (ionic) media, and the results compared with the experimental results.

４．研究成果

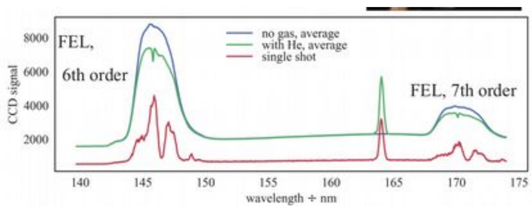
1) Development of a gas cell for creating a high density of neutral atoms and ions (see publication 2). In this approach, a pulsed gas valve was used to create a pulse of gas (duration hundreds of microseconds) in a high vacuum (<1E-4 Pa) chamber connected to a free-electron laser beamline. The pulsed nature of the source allows high densities to be reached without compromising the high vacuum requirement. Further, a 2-stage gas cell was used to increase differential pumping, reaching estimated instantaneous number densities of up to 1E25 atoms per cubic metre. Using free-electron laser pulses with photon energies greater than the ionisation potential of the atoms used, high densities of singly-charged ions could be instantaneously created and studied with a single pulse.

2) Observation of Raman scattering and free-induction decay following excitation of neutral helium with a free-electron laser. Using a grazing-incidence extreme-ultraviolet spectrometer (Shinku kogaku), and an X-ray streak camera (Hamamatsu photonics), the propagation of free-electron laser pulses through dense samples of neutral helium gas was studied. Example results are shown in the figure below (unpublished, but in preparation).



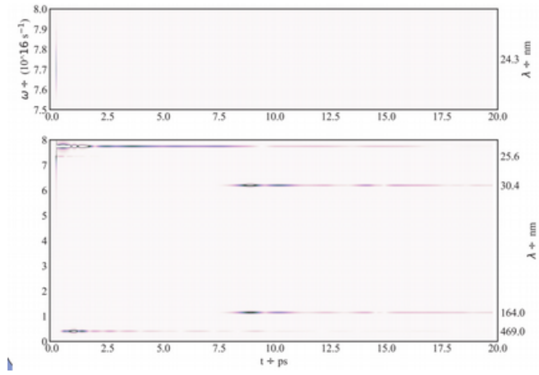
Streak camera traces of incident FEL pulses (black) and resonant emission at different number densities.

3) Observation of 'superfluorescence' from hydrogen-like helium (He^+). By using free-electron laser pulses at a photon energy of 51 eV, we were successful in ionising and exciting sufficient helium atoms in a single pulse to observe superfluorescence from excited 4p helium ions. Strong, directional emission was observed at 469 nm (n=4 to n=3) and also 164 nm (n=3 to n=2). To our knowledge, the emission at 164 nm, if indeed superfluorescence, represents the shortest wavelength superfluorescence reported to date.



Intense, directional emission observed at 164 nm (n=3 to n=2 transitions of He^+) following ionisation end excitation of helium to He^+ (4p)

4) Numerical simulations (see publications 3, 4). Based on an approach outlined in the literature (Marskar and Osterberg, Optics Express **19**(18) 16784 (2011)), multi-level simulations were developed to treat the interaction of ensembles of partially-coherent free-electron laser pulses with samples of atomic and ionic helium at the densities used in the experiments. The figure shows results of a simulation of the propagation of a free-electron laser pulse at a central photon energy of 51.0 eV through a dense sample of helium ions. The plot shows a rolling Fourier transform of the electric field at the exit of the atomic medium, and reveals free-induction decay at the resonant excitation frequency, Raman scattering from non-resonant dipole-allowed levels, and cascade superfluoresence at wavelengths of 469 nm and 164 nm, in agreement with the results of the experiments performed at SACLA. These results are being prepared for publication.



Rolling Fourier transform of the output electric field following propagation of a free-electron laser pulse through a dense sample of helium ions (see text for details).

5) Further development of an electron-beam ion trap suited to studies at free-electron laser and synchrotron lightsources. This work was carried out in collaboration with a group centred at the Max-Planck Institut fur Kernphysik, and is summarised in publication 1 (Micke et al, Rev. Sci. Instrum. **89** (6) 063109 (2018)). Conventional electron-beam ion traps are the size of a large laboratory, and use superconducting magnets and high energy, high-current electron beams to create and trap ions of almost any charge state in a hot, electron-dense plasma-like state. While use of such a trap has been demonstrated at synchrotron radiation and free-electron laser beamlines, the goal of this part of the research is to develop a more compact design of trap. The use of permanent magnets drastically reduces the size

and cost requirements, at the expense of the ultimate charge states which can be reached. Four new compact, permanent-magnet, room-temperature electron beam ion traps have been developed and tested (one at the applicant's institute in Japan, the others in Germany). The traps have demonstrated the production and trapping of ions with charge states of up to +29. One of the traps developed in Germany has been already used successfully at a synchrotron radiation beamline to study the photo-ionisation of highly-charged ions. The trap constructed in Japan uses specially-designed in-vacuum polepieces to reach higher magnetic fields, and is being prepared to be used at a free-electron laser beamline.

５．主な発表論文等
（研究代表者、研究分担者及び連携研究者には下線）

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