様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　５ 年　６ 月 ２６ 日現在

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研究課題名（和文）Superfluorescence, free-induction decay, and four-wave mixing: experimental and numerical studies of the propagation of free-electron laser pulses through dense atomic/ionic media

研究課題名（英文）Superfluorescence, free-induction decay, and four-wave mixing: experimental and numerical studies of the propagation of free-electron laser pulses through dense atomic/ionic media

研究代表者

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交付決定額（研究期間全体）：（直接経費）　　3,300,000円

研究成果の概要（和文）：本研究の目的は自由電子レーザーを用いた短波長（極端紫外領域~軟X線領域）における量子光学効果の実現である。研究の焦点は光源のコヒーレンスと物質内のダイナミックスから発展するコヒーレンス両方に影響される超蛍光という1950年代から知られている現象であって、その現象のEUV領域の実現と測定をテーマにしている。今までは間接的に測定できたEUV領域の超蛍光（波長164）の直接検出（時間次元の検出）と、数値計算との比較を目指し、また、より短い波長領域への道を開けることが研究開始当時の計画である。自由電子レーザーを用いた実験を行い、164 nmにおける超蛍光の時間構造と数値計算との比較に成功した。

研究成果の学術的意義や社会的意義
学術的:X線領域における量子効果は様々な光源と様々な物質で研究の対象となっている。本研究ではその光源の一つである自由電子レーザーのコヒーレンス性に敏感な現象に注目し、今後の光源利用、手法開発に貢献できた。
社会的:「量子」をキーワードとした技術開発、手法開発は今後大きなテーマとなる。本研究ではその開発のためのプローブである自由電子レーザー光源の量子性の一つであるコヒーレンスと物質のコヒーレンス性に注目し、今後の研究方針や研究基盤に貢献できた。

研究成果の概要（英文）：The scientific goal of the main focus of this research is the use of free-electron laser light sources to extend quantum optical effects well-established at visible wavelengths to the extreme-ultraviolet and shorter. Of particular interest is the phenomenon of "superfluorsecence", which is sensitive to the coherence of the light source used and also to coherence developed from dynamics within the medium. In earlier work we indirectly observed this phenomenon at short wavelengths (164 nm, 30.4 nm), and the goal was to directly observe the superfluorescence emissions in the time domain and compare with numerical simulations. A further goal is to explore routes to extending the observations to even shorter wavelengths.
We were successful in observing the 164 nm superfluorescence in the time domain, and comparing it with numerical simulations.

研究分野：原子分子物理学

キーワード：自由電子レーザー　超蛍光　ヘリウム　コヒーレンス　量子光学

１．研究開始当初の背景

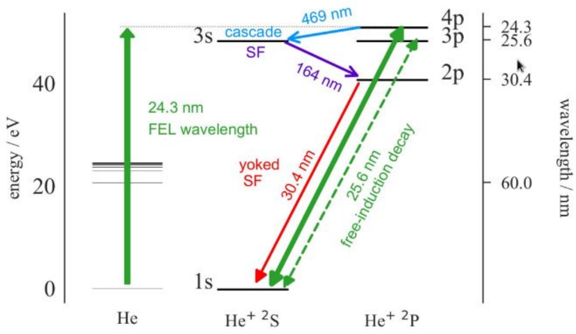

Figure 1 Level diagram of He+ (see text)

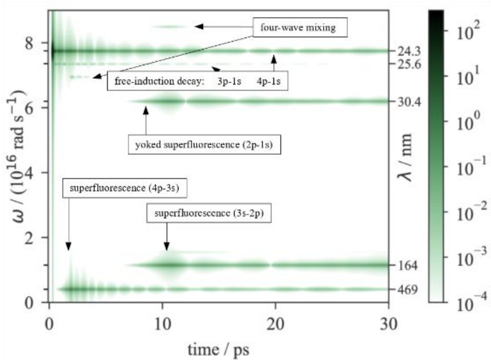

Figure 2 Simulation results (see text)

This research builds on previous work observing superfluorescence and related phenomena using pulses of radiation from a free-electron laser to create incoherently-excited population inversions in dense samples of helium gas[1–4]. Using the shorter wavelengths available at SACLA BL1, we successfully demonstrated that superfluorescence could also be generated in a dense sample of helium ions, where the free-electron laser pulses serve to both ionize the neutral atoms and resonantly excite them. In experiments with the free-electron laser wavelength tuned to 1s-4p excitation (figure 1) we observed strong, directional emission at wavelengths of 469 nm (4p-3s) and 164 nm (3s-2p), which can be attributed to classical cascade superfluorescence on these two transitions. The superfluorescence nature of the transition at a wavelength of 469 nm was confirmed by detecting the time structure of the emission using a fast photodiode. We also observed strong directional emission at wavelengths of 24.3 nm (the same wavelength as the excitation wavelength), 25.6 nm (3p-1s) and 30.4 nm (2p-1s), which are not expected in the classical superfluorescence picture since no population inversion exists on these transitions. The results of the simulation (figure 1) suggested that the emissions at 24.3 nm and 25.6 nm can be attributed to free-induction decay on these transitions, and that the emission at 30.4 nm is *yoked* superfluorescence, and related to the partial coherence of the excitation. However these shorter wavelength transitions could not be observed in the time domain due to the unavailability of appropriate detection techniques. Further, comparison with simulation was hampered by a lack of knowledge of the density and distribution in space of the target atoms/ions.

２．研究の目的

The potential observation of coherent quantum optical effects using a SASE (self-amplified spontaneous emission) source is of great interest to study since these sources are more prevalent than fully-coherent sources at soft X-ray wavelengths, and unavailable at even shorter wavelengths. Superfluorescence itself is also of great interest for its intensity and directionality, with potential applications for studying very weak transitions (due to the effective amplification it offers), and as such is even being considered as a potential detector for dark matter. The goals of this research were to attempt to create overlap between simulation and experiment, confirm our results by carrying out time-resolved detection of the emissions mentioned above, and also investigate routes to extend our techniques and observations to even shorter wavelengths. As such we had the following goals at the start of this project:

1) Time-resolved detection of the short wavelength emissions (164 nm and shorter), and developing the simulation techniques to closer match experimental parameters.
2) Developing a method to determine sample number density shot-by-shot
3) Exploring ways of extending the work to even shorter wavelengths using higher-order radiation from the free-electron laser, and even higher sample densities.

３．研究の方法

In pursuit of the goals mentioned in section 2, the following research directions were taken.

1) Time-resolved studies at the wavelengths relevant to this work are much more difficult than at visible wavelengths, due to the limited availability of optical components at short wavelengths, and the necessity of operating under vacuum conditions. In earlier work we

attempted to combine a grazing-incidence monochromator with an X-ray streak camera to study wavelength-dispersed time resolved emission. However since both of these techniques are inherently low-yield, the combination proved difficult to align and the signal too weak to carry out more than proof-of-principle experiments. In this work we pursued two approaches: 1a) using a fast photodiode and a wavelength filter to study the 164 nm emission, and 1b) replacing the CCD camera detector of the grazing-incidence spectrometer with a fast MCP detector and a slit. The obtained results were compared with simulations.

2) Various approaches were considered for studying the instantaneous gas density, and two interferometry approaches were tested offline. By passing a laser beam through the gas cell and detecting time-resolved interference patterns we envisage that a shot-by-shot diagnostic can be developed for the next series of experiments using the free-electron laser source..

3) For shorter wavelength studies we first investigated using an absorption technique the feasibility of using higher-orders of the free-electron laser radiation. Since superfluorescence requires number densities high enough that wavelengths of emission are comparable to atom spacing, shorter wavelengths in general require higher densities. With a gas sample our technique is approaching the limit, and one direction we explore is using a cluster or droplet sample, where the density approaches that of the bulk but the convenience of a gas sample remains.

４．研究成果

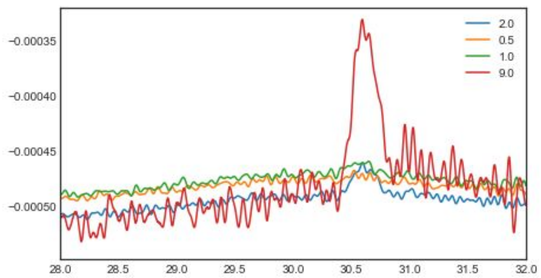

Figure 3 Fast UV photodiode traces at different sample densities

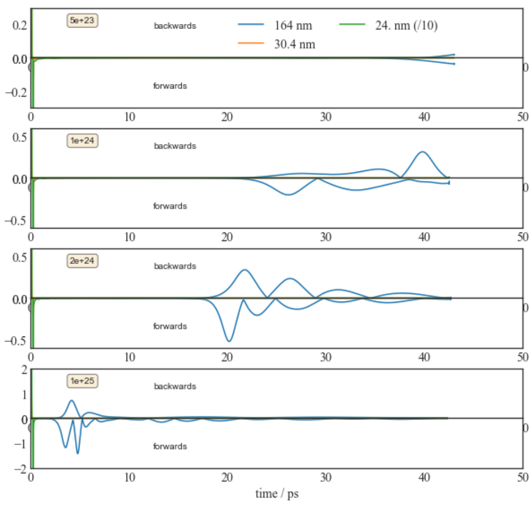

Figure 4 Simulation for 1s-3p excitation

1) The first experiment we attempted used a fast UV photodiode (alphalas systems) with a nominal 170 nm – 1100 nm range but some sensitivity at 164 nm. Results of traces with a 164 nm bandpass filter (thor labs) positioned in front of the detector are shown in figure 3. Corresponding simulations are shown in figure 4. The simulations show that 164 nm emission should be observed in both forward and backwards directions, with the delay with respect to excitation reducing with increasing number density. Figure 3 shows traces recorded at 4 different number densities (the legend shows the pressure in atmospheres of the gas behind the pulsed nozzle, which is proportional to number density). The x-axis is time in ns (prior to determination of time zero, which is around 30.5 ns). While it is clear that the emission is pulsed in nature and increases in intensity with increasing gas pressure, the sensitivity of the detector was not sufficient to observe the time delay. In a further experiment we returned to using the grazing-incidence spectrometer, but replaced the CCD detector with a fast MCP detector. Positioning the grating so that wavelengths of 164 nm were reflected centrally through a slit in front of the detector, the time structure of emitted pulses could be detected (figure 5). This figure shows not individual pulses, but the distributions of the peak positions for hundreds of pulses recorded at each number density (labels show the gas pressure behind the pulsed nozzle). The distribution labelled '0.0 MPa' corresponds to background scattered radiation and serves as an instrument function. For 3p excitation (upper panel) it is clear that there is a short delay between excitation and emission, clearly confirming that emission at this wavelength is indeed superfluorescence. At 4p excitation this delay is even more apparent (lower panel), and a comparison of the simulations for 4p excitation (figure 6) and 3p excitation (figure 3) explain the observations. Within the limited experimental time available it was not possible to extend

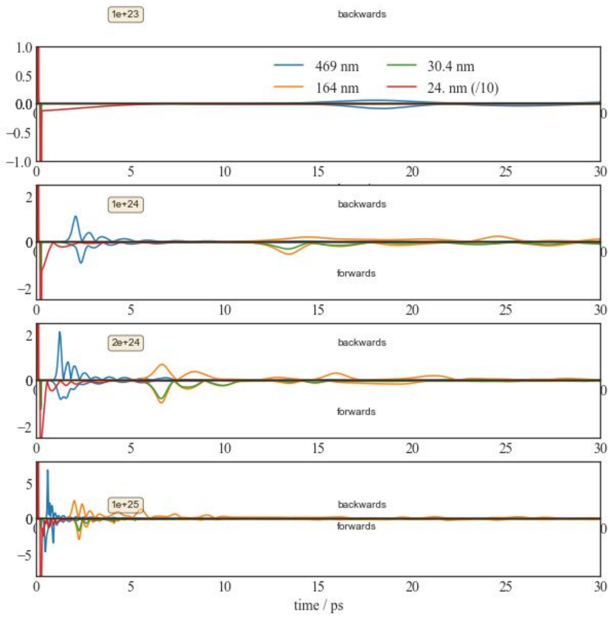

Figure 5. Simulations for 1s-4p excitation

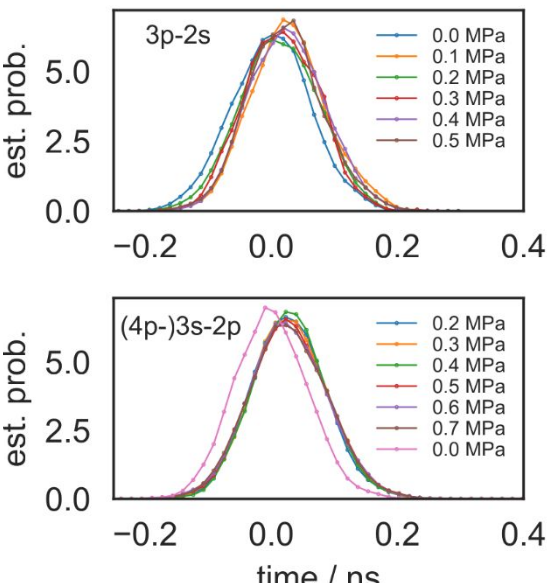

Figure 6. Distributions of arrival times of pulses recorded using a fast MCP detector

the measurements to the other wavelengths of interest (24.3 nm, 30.4 nm), but the experimental method has been demonstrated, and with the addition of an adjustable slit and reproducible wavelength selection to the spectrometer setup it is anticipated that future experiments will be successful in this regard.

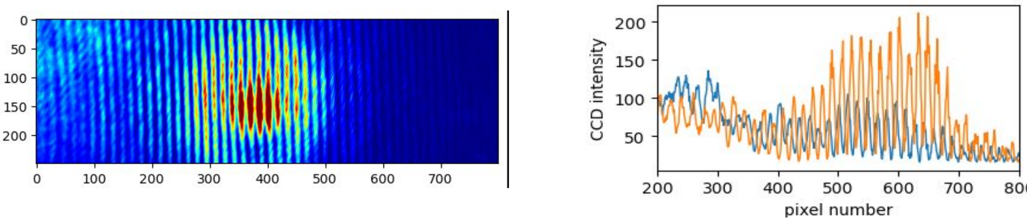

Figure 7. Interference patterns (left) and lineouts from the Mach-Zender interferometer

2) Using a Mach-Zender setup and a laser diode we continued to develop a shot-by-shot method for determining number density in the gas cell. Figure 7 shows an image of an interference pattern from a laser beam split before entering the vacuum chamber and recombined afterwards, with one arm of the interferometer able to be positioned just above the gas jet. On the right are lineouts of the intensity at different positions – comparison of these on a shot-by-shot basis will allow for shot-by-shot estimates of number density of the sample which the free-electron laser passes through. A new gas cell will be constructed in the future to allow this.
3) Using the grazing-incidence spectrometer and the same gas cell used for the superfluorescence experiments we were able to carry out near-edge x-ray absorption fluorescence spectroscopy for the argon 2p and CO_2 C 1s and O 1s core ionisations. This demonstrates that intensity at these wavelengths (photon energies around 250 eV, 290 eV, and 530 eV) are high enough to observe absorption of the beam – a necessary requirement for superfluorescence at these wavelengths.
For sample development, we continued to consider the use of a cluster or droplet beam to reach number densities approaching the bulk. Figure 8 shows example mass spectra using a residual gas analyzer (SRS RGA 100) and a pulsed helium nozzle cooled to a temperature of 5 K. By recording the output of the electron multiplier using an oscilloscope the pulsed beam (upper) could be separated from background gas (lower). Signal is observed and multiples of the helium mass of 4, with magic numbers observed at e.g. N=7. At low temperatures the significant increase in intensity of the N=4 peak (m/q = 16) is indicative of large droplets, and we estimate that these conditions correspond to sizes of around 10^7 atoms. With an N of 10^7 and bulk spacing, we hope that superfluorescence or related effects can be observed. Compared to the atomic (gaseous)

situation, the evolution of effects such as superfluorescence is greatly complicated by the increased number of degrees of freedom in the sample, for example phonon dynamics.

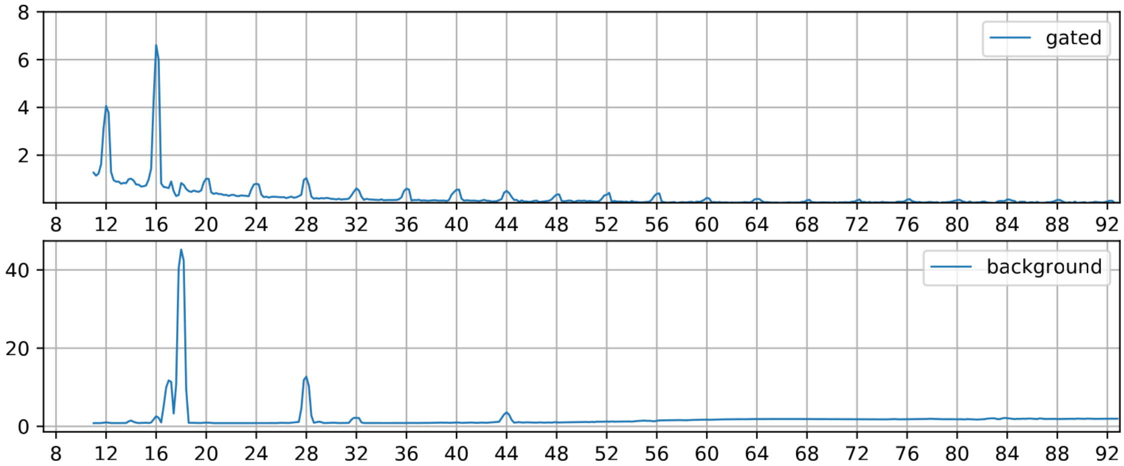

Figure 8. Mass spectra of pulsed droplet beam. x-axis: m/q. Pulsed beam (upper), background gas (lower)

５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　1件／うち国際共著　0件／うちオープンアクセス　1件）

１．著者名 Hiroshi Iwayama, Masanari Nagasaka, Ichiro Inoue, Shigeki Owada, Makina Yabashi, James Harries	４．巻 10,7852
２．論文標題 Demonstration of Transmission Mode Soft X-ray NEXAFS Using Third- and Fifth-Order Harmonics of FEL Radiation at SACLA BL1	５．発行年 2020年
３．雑誌名 Applied Sciences	６．最初と最後の頁 1-7
掲載論文のDOI（デジタルオブジェクト識別子） 10.3390/app10217852	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 －

〔学会発表〕　計9件（うち招待講演　4件／うち国際学会　3件）

１．発表者名 James Harries, Arisa Iguchi, Hiroshi Iwayama, Susumu Kuma
２．発表標題 Photoionisation and excitation of dense samples of helium gas using an XFEL: superfluorescence at EUV wavelengths
３．学会等名 IWP-RIXS 2022（国際学会）
４．発表年 2022年

１．発表者名 James Harries, Arisa Iguchi, Susumu Kuma
２．発表標題 A new superfluid liquid helium droplet machine
３．学会等名 原子衝突学会第47回年会
４．発表年 2022年

１．発表者名 ハリーズ　ジェームズ
２．発表標題 Short-wavelength superfluorescence and related phenomena observed at SACLA
３．学会等名 第35回日本放射光学会年会・放射光科学合同シンポジウム（招待講演）
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1．発表者名 ハリーズ　ジェームズ
2．発表標題 原子からの超蛍光
3．学会等名 日本物理学会 第77回年次大会（招待講演）
4．発表年 2022年

1．発表者名 J R Harries
2．発表標題 世界最短波長「超蛍光」の観測
3．学会等名 レーザー学会学術講演会第41回年次大会（招待講演）
4．発表年 2021年

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2．発表標題 自由電子レーザーによるヘリウムガスにおける超蛍光の観測
3．学会等名 第 21 回レーザー学会東京支部研究会
4．発表年 2021年

1．発表者名 J R Harries
2．発表標題 Observation of extreme ultraviolet superfluorescence from helium atoms ionised and excited using soft x-ray free-electron laser pulses
3．学会等名 XXXIst International Conference on Photonic, Electronic, and Atomic Collisions（招待講演）（国際学会）
4．発表年 2019年

1．発表者名 J R Harries
2．発表標題 Development of a pulsed liquid helium droplet source for use at synchrotron and free-electron laser sources
3．学会等名 SACLA Users' Meeting 2019（国際学会）
4．発表年 2019年

1．発表者名 J R Harries
2．発表標題 Extreme ultraviolet superfluorescence from helium ions
3．学会等名 第35回化学反応討論会，第35回化学反応討論会実行委員会
4．発表年 2019年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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6．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考
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7．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

8．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関