様式 C-19

科学研究費補助金研究成果報告書

平成 22 年 3 月 11 日現在

研究種目：若手研究（B）
研究期間：2007～2008
課題番号：19740254
研究課題名（和文）寿命別蛍光・光イオンコインシデンス分光を用いた原子・分子の内殻光励起研究
研究課題名（英文）Atomic and molecular inner-shell photo-excitation studied using a photo-ion : lifetime-resolved fluorescence coincidence technique.
研究代表者
HARRIES, James （ハリーズ・ジェームズ）
日本原子力研究開発機構・量子ビーム応用研究部門・研究職
研究者番号：30416383

研究成果の概要

本研究では原子・分子の放射光内殻励起過程を調べるために新しい実験法を開発した。中性粒子（原子および光子）の検出およびイオンとの同時検出によって水分子の酸素原子の内殻励起後の分子の解離について新しい情報が得られた。中性水素原子が生成する場合、その原子の励起状態分布についておよび残りの OH イオンの解離過程について情報を得て、議論ができた。詳細結果については投稿論文として準備中である。

交付額

（金額単位：円）

	直接経費	間接経費	合　計
2007 年度	1,900,000	0	1,900,000
2008 年度	1,200,000	360,000	1,560,000
年度			
年度			
年度			
総　計	3,100,000		3,460,000

研究分野：数物系科学
科研費の分科・細目：物理学・原子・分子・量子エレクトロニックス・プラズマ
キーワード：原子・分子

１．研究開始当初の背景

(1) Prior to the start of this research a preliminary study of inner-shell photo-excitation of neon had shown the potential of lifetime-resolved fluorescence spectroscopy for studying these processes. However due to the large number of final states potentially reachable, it seemed worthwhile to attempt fluorescence detection in coincidence with photo-ions.

(2) At the start of this research the principal investigator was attached to beamline 27SU at SPring-8, which is ideal for the study of inner-shell excitation processes in atoms and small molecules.

(3) At other research groups inside and outside of Japan there was interest in the success of this technique.

(4) It was also known that near the N (1s) threshold in N_2 neutral, long-lived, excited N atoms could be produced following excitation/ionisation.

２．研究の目的

(1) The ultimate aim of the work was to perform detailed studies on the photo-excitation, ionization, and dissociation processes in atoms and small molecules.

(2) The first step was to study the total fluorescence and neutrals yield in the inner-shell excitation region for a range of species (starting with Ne and H_2O) to select suitable cases.

(3) The next step was to attempt fluorescence-ion coincidences.

(4) During steps 2 and 3 it was found that as well as fluorescence-ion coincidences, neutral (excited) particles: ion coincidences should also prove a useful technique to develop.

３．研究の方法

(1) The key part of the technique is the SPring-8 beamline BL27SU, which is one of the most suitable beamlines in the world for the studies here, providing both high-resolution and high flux in the photon energy region of interest

(2) The technique also makes use of the pulsed nature of the synchrotron source.

(3) A simple apparatus consisting of multi-channel plate detectors was constructed and installed at the beamline to detect total fluorescence and neutral particle yields. All signals were detected in relation to the timing structure of the synchrotron radiation, recording the time intervals between detection of a particle and the next pulse of light. SPring-8 offers several operation modes with different time structures, and “D” or “E” modes were chosen, which offer the longest pulse separations.

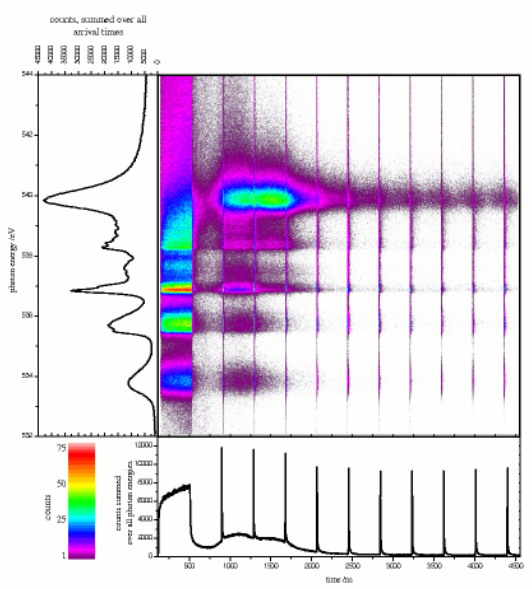
(4) An ion time of flight tube was designed and constructed to allow for time-of-flight ion detection.

(3) A Lyman alpha filter was purchased to allow the selective detection of (2p->1s) hydrogen atom fluorescence resulting from the decay of inner-shell excited H_2O molecules.

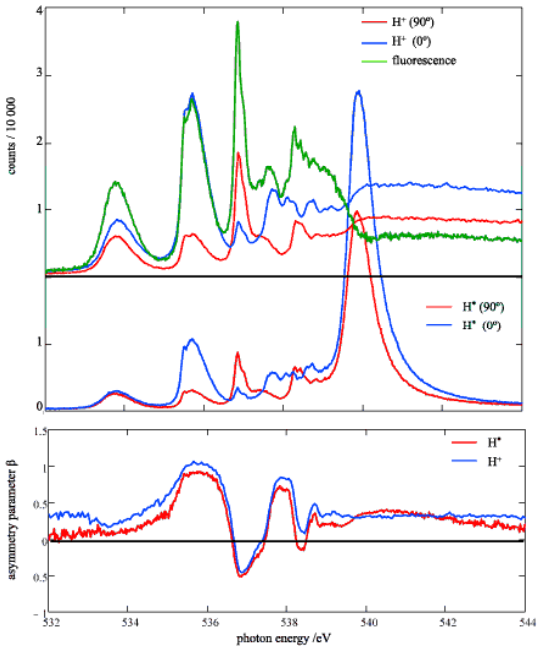
(4) Data analysis techniques were developed for analyzing the fluorescence lifetimes, and also for distinguishing between false and true coincidences.

４．研究成果

(1) Total neutral yield.



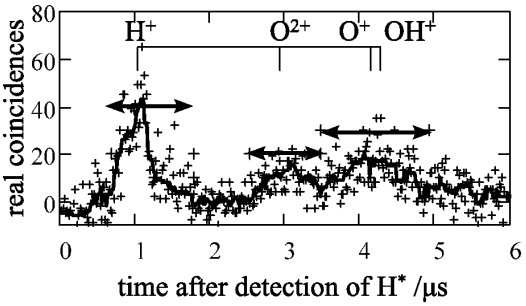
The figure above shows the time-resolved, total neutral yield in the region of O (1s) excitation of water. The x-axis is the time between the detection of a particle and the next pulse of synchrotron radiation; the y-axis is the incident photon energy. The panels to the left and below show sums over the perpendicular direction, revealing the total neutral yield as functions of photon energy (left) and flight time (bottom). Fluorescence photons travel at the speed of light, and reach the detector in nanoseconds. Neutral fragments arrive in microseconds, allowing these two channels to be easily separated. Resolving the yield in the time axis in this manner in these experiments is unique to this work. Different time regions can



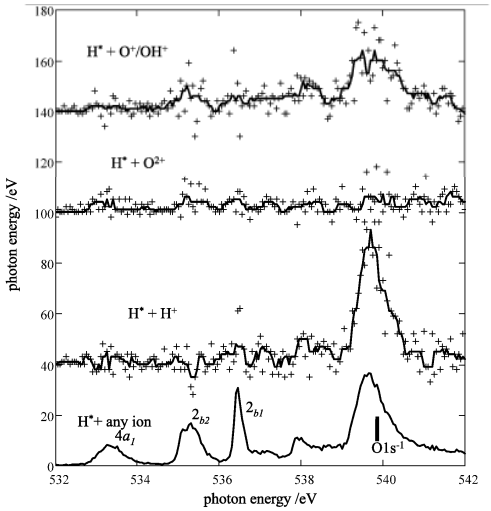
be analysed to provide different information. For example total fluorescence and total neutral yield can be separated, as shown in figure 2. Further, analysing the lifetimes of the fluorescence decays leads to information on the final H atom states, which have different lifetimes. Analysing the flight-times of neutral atoms leads to information on the dissociation process, since different processes and final states lead to different energy releases and particle speeds.

(2) The second figure shows the separated neutrals and fluorescence yields, resolved by the timing technique. Also shown are ion yields for comparison. Neutral, excited H atoms are produced following Auger decay and subsequent dissociation. In particular a large peak is seen at the O (1s) threshold for neutral atom production whereas only a step increase is seen in the ion yield. This provides new information, and a publication is in preparation for submission to the Journal of Physics B.

(3) It was discovered that the neutral atom yield was much higher than the fluorescence yield, and consequently neutral:ion coincidence experiments were

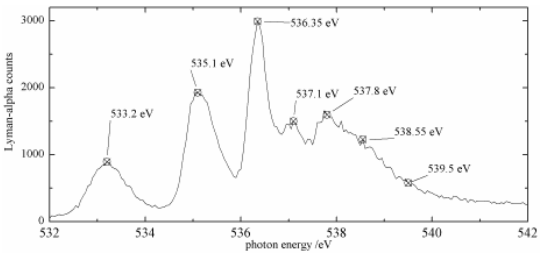


performed. A sample spectrum is shown above. Here the detection of a neutral excited H^* fragment triggers the detection setup, which is stopped by the detection of an ion in the other detector. Careful treatment of "false" coincidences, where an ion from a different event stops the timer, must be carried out. The time difference depends on the flight time, and thus the species of ion. It can be seen that H^+, O^{2+}, and O^+ or OH^+ ions are detected, shedding light on the dissociation process. For N (1s) excitation in N_2 leading to excited N atoms the counterpart can only be a N+ ion. For H_2O however a three-body breakup occurs, and the analysis is more



complicated. Scanning the incidence photon energy and extracting the coincidence pairs shown by arrowed lines in the time-of-flight figure leads to the spectra shown above. While (H^*+H^+) seems to be the dominant channel, it is only seen at threshold, where the initial excitation leads to a Rydberg state. This pair is presumed to be created when the OH+ fragment with an inner-shell hole undergoes dissociation into O+H^+. The neutral O atom is unobserved. (H^*+OH^+) is seen at both threshold and also at valence-state excitation energies. The (H^*+O^{2+}) channel is presumably created when the inner-shell hole OH^+ ion undergoes Auger decay and dissociates. It is unclear at which incident photon energies this process occurs, but it does not occur preferentially at threshold. A detailed analysis of these results will be published in the Journal of Physics B.

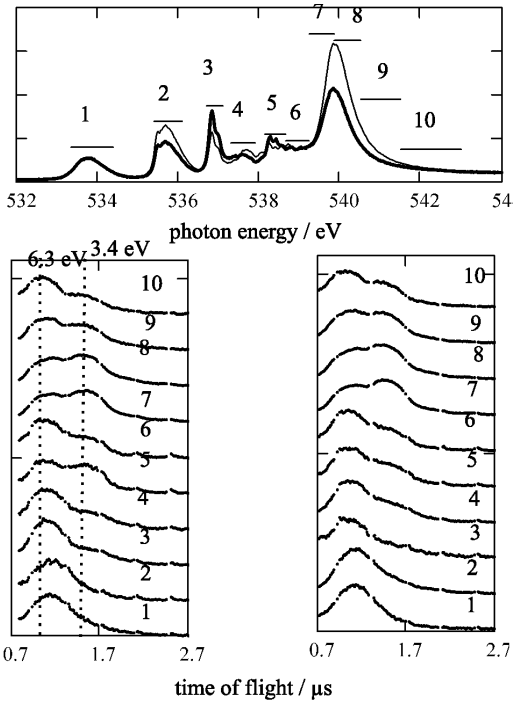
(4) While the fluorescence yield was not high enough to allow coincidence experiments to be performed with the current apparatus, the use of a Lyman alpha filter allowed a detailed analysis of the

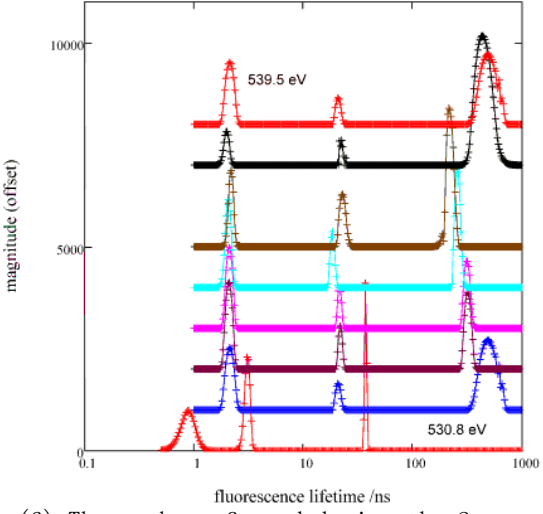


final state neutral H atom populations following dissociation. The figure above shows the Lyman alpha yield recorded in the same energy region as is shown in figure 2. The figure below shows the fluorescence

lifetimes revealed by an analysis of the time-resolved fluorescence spectra recorded at the points labelled in the upper figure. The conventional analysis technique is to fit the data to a sum of Gale's functions (an exponential decay convolved with a Gaussian "instrument function"), and this works well when there are a small number of discrete lifetimes. When there are a larger number of lifetimes, or distributions of lifetimes, a better approach is to use a grid of lifetimes spaced equidistant in logarithmic space. Results from using this technique are shown in the second figure above. Particularly striking is the increase in long lifetimes close to the O 1s threshold at 540 eV: this is due to the production of long-lived Rydberg states. A detailed analysis is under preparation for publication.

(5) As mentioned in (2), analysis of the times-of-flight of the neutral H* particles also provides information on the dynamics of the breakup of the molecule. The figure below shows the time-of-flight spectra of H* atoms recorded at several incident photon energies, shown by horizontal lines above the total H* yield traces in the top panel. The spectra on the left for detection parallel to the polarisation vector of the incident radiation and those on the right for detection perpendicular are very similar, but there is a strong incident energy dependence, with roughly three different peaks discernible. This work is also under preparation for publication.





(6) The work performed during the 2 years has demonstrated the applicability of the technique. Apparatus developments would allow for lifetime:ion coincidence experiments to be performed. Improvements in the lifetimes analysis technique will also lead to more information on the final state distribution.

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）

〔雑誌論文〕（計 0 件）

〔学会発表〕（計 10 件）

(1) HARRIES, James "Metastable fragment and VUV fluorescence yield following O 1s excitation of H2O", "9th European Conference on Atomic and Molecular Physics", June 2007, Heraklion (Greece).

(2) HARRIES, James. “寿命選別測定法で観測する分子の解離ダイナミックス”.” BL27SU の現状と今後の展開”, October 2007, SPring-8 (Japan).
(3) HARRIES, James “Neutral H* production following O (1s) excitation /ionization of gas-phase H2O”. “第 21 回日本放射光学会年会”, January 2008, Kusatsu (Japan).
(4) HARRIES, James “酸素 01s の内殻励起による水のイオン化解離ダ イナミクス”. “第２４回科学反応討論会”. June 2008, Sapporo (Japan).
(5) HARRIES, James “Production of neutral particles following O 1s excitation of H2O: H* yield and kinetic energy, and ion-H* coincidence studies.” “The International Workshop on Photoionization 2008”. June 2008, Sala (Sweden).
(6) HARRIES, James “Metastable H atoms from O 1s excitation/ionisation of H_2O”. “21st International Conference on X-ray and Inner-Shell Processes”. June 2008, Paris (France).
(7) HARRIES, James “気相水分子の O1s 励起 中性高励起フラグ メントと寿命別蛍光”. “日本物理学会秋季大会”. September 2008, Morioka (Japan).
(8) 下條竜夫,“間分解ケイ光測定によるアルゴンクラスター崩壊過程の研究”. “第 2 回分子科学討論会”.September 2008, Fukuoka (Japan).
(9) 國分美希“酸素 O1s の内殻励起による水の光解離ダ イナミクス”. “第２回分子科学討論会”. September 2008, Fukuoka (Japan).
(10) HARRIES, James “Time-Correlated Single-Photon Counting experiments for atomic, molecular, and cluster photo-excitation studies.”. “Pf 研究会「PF リングのトップアップ・シングルバンチ運転利用研究と今後の発展について」”. November 2008, Tsukuba (Japan).

〔図書〕（計 0 件）
○出願状況（計 0 件）

○取得状況（計 0 件）

〔その他〕

６．研究組織
(1)研究代表者

HARRIES, James （ハリーズ・ジェームズ）
日本原子力研究開発機構
量子ビーム応用研究部門
放射光表面・薄膜創製研究グループ
研究者番号：30416383

(2)研究分担者（なし）

(3)連携研究者（なし）

(4)研究協力者

① 下條 竜夫（げじょう たつお）
兵庫県立大学
大学院
物質理学研究科
准教授

② 東 善郎（あずま よしろう）
上智大学
理工学部
物質生命理工学科
教授

③ SULLIVAN, James
（サリバン・ジェームズ）
Australian National University
RSPhysSE
Australian Research Fellow