

時間相関単一光子計数法を用いた原子、分子、クラスターの光励起実験

Time-Correlated Single-Photon Counting experiments for atomic, molecular, and cluster photo-excitation studies

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The time-correlated single photon counting technique most commonly makes use of pulsed laser excitation sources. It has widespread applications [1], particularly in the imaging of biological materials, where dyes which fluoresce with particular lifetimes are used to stain particular regions of a sample. Compared to laser radiation single-bunch synchrotron radiation has the disadvantages of lower intensities, broader pulses, non-variable pulse structure, non-portability, and high cost. However in the VUV-SX energy region synchrotron radiation offers the ability to study systems and processes otherwise inaccessible.

The core technique is to detect fluorescence photons in coincidence with a signal corresponding to the time structure of the incident radiation. Provided the incident pulse width is narrow enough and the pulse separation is long enough, lifetime(s) of the fluorescence decay processes can be determined. The initial populations of the fluorescing states can also be determined. These populations can then be studied as other parameters are varied – for example the incident photon energy, a technique referred to as lifetime-resolved fluorescence spectroscopy (LRFS).

The valence/Rydberg fluorescence decay processes in atoms and molecules have lifetimes on the scales of picoseconds to microseconds. Synchrotron radiation consists of short pulses (1-100 ps) separated by anything from 2 ns to 5 μ s depending on the size of the storage ring and the mode of operation, enabling the study of many different systems. The simplest measurements involve using synchrotron radiation to excite particular states and then detecting the radiation emitted as they decay to the ground state. Systems studied include the helium singly-excited Rydberg states [2], and an extension of the technique has been used to study quantum beats in the rare gas atoms [3]. This technique was also used to provide the first direct measurements of the

lifetimes of the helium doubly-excited states [4, 5].

The LRFS technique requires a system with a small number (2-10) of known lifetimes, which are ideally not similar, and was first successfully applied at the PF using synchrotron radiation to determine the populations of the $3s$, $3p$, and $3d$ excited states of hydrogen atoms produced following dissociation induced by the absorption of 73.4 nm wavelength photons [6]. The next successful application was to He^+ fluorescence, and the partial cross-sections for the production of the $2s$, $2p$, (and $3s$, $3p$) $3d$ ion states were determined in the $\text{He } 3nl'$ (and $4nl'$) double excitation energy region [7, 8]. These experiments used (non top-up) single-bunch operation at the PF and two-bunch operation at the ALS. More recently the technique has been applied at SPring-8 in the SX region, where inner-shell excitation occurs. For the inner-shell excitation of argon clusters [9], cluster fluorescence lifetimes were found to differ from atomic ones. New information can be obtained by considering specific fluorescence decays from final ion states following Auger decay, and the example of Ne $1s$ excitation will be discussed. The excited H system is one which is particularly well-suited to LRFS studies, and a recent study of the production of excited H fragments following $\text{O}(1s)$ excitation/ionization of H_2O will be presented.

A comparison of studies using different synchrotron radiation sources and operation modes will be presented, with the main conclusion that single-bunch, top-up operation offers exciting opportunities to develop this technique and its applications.

References

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