

## 1-1 Lifetime-resolved Fluorescence Spectroscopy as a Sensitive Tool for Atomic and Molecular Excitation/Ionisation Processes

Processes on the atomic and molecular scale often lead to the production of decay products in both ground states and also in excited states. Knowledge of these reactions, the distribution of final states, and the time scales on which they occur is fundamental to our understanding of chemical reactions and processes in systems ranging from plasmas in laboratories to stars in distant galaxies. The most widely used technique for these studies is analysis of the energy of the electrons produced when a system decays. When the input of energy into the system (the energy of the photon in the case of synchrotron radiation experiments) is known, determining the energy of the electron gives information about the state of the decay products. Alternatively, the radiation emitted when excited states decay can be analysed. Conventionally, this radiation is dispersed, and the energy of the photons emitted determined. In this work we analyse the emission not in terms of its energy distribution, but its time dependence. Since different excited states have different lifetimes, for simple systems this approach can differentiate between final states which are practically degenerate in energy. The technique can be extended to more complex

systems by combining time-resolved detection with energy-resolved detection of either the decay photons or the electrons, but here we concentrate on photo-excitation/ionisation of helium, since the excited states of the ion are a simple and very well known system. Similar studies have also previously been carried out at the PF on molecular hydrogen [1]. In the photoionisation of helium, an energy region of particular interest is that where ionisation can take place via the resonant excitation of doubly-excited states. Here we present results in the "3lnl" region where one electron is excited to an  $n=3$  state, one to a Rydberg state. This unstable system then autoionises, leaving an ion in a state with  $n=1,2$ , or 3. For the excited ion states, we detect the radiation emitted, and determine which final state was produced by analysing the lifetime of the emission. To allow this, single-bunch mode operation of the PF storage ring is essential, providing 628 ns between each photon pulse. (In general, a spacing long compared to the longest lifetime under study is essential for this technique.) The results presented here were obtained at BL-16B. Fig. 1 shows an example of the time-resolved fluorescence signal in the helium 3lnl' excitation region, and demonstrates that the signal consists of four components due to the decay of states with different lifetimes. The lifetime-resolved fluorescence spectroscopy technique consists of recording these spectra whilst scanning the incident photon energy, and extracting the separate lifetime components at each step. The results

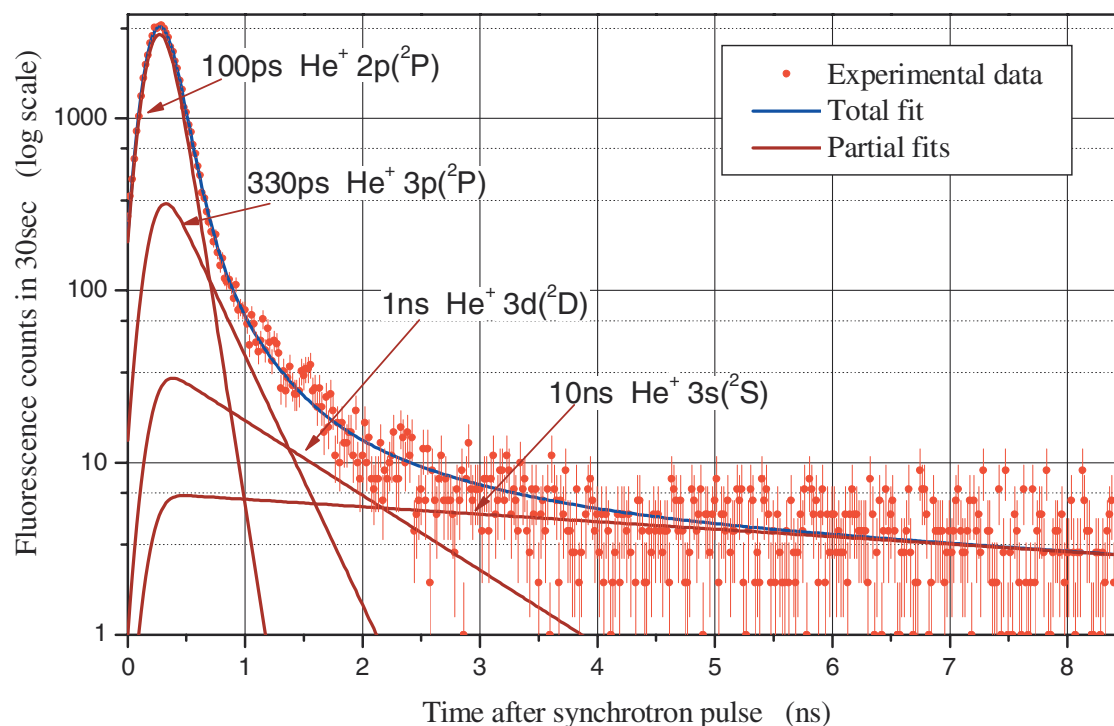


Figure 1  
An example of a time-resolved fluorescence spectrum in the He 3lnl' excitation region. The overall fit (blue) consists of the sum of 4 separate lifetime components (brown) and a constant background due to fluorescence of a 2ms lifetime state.

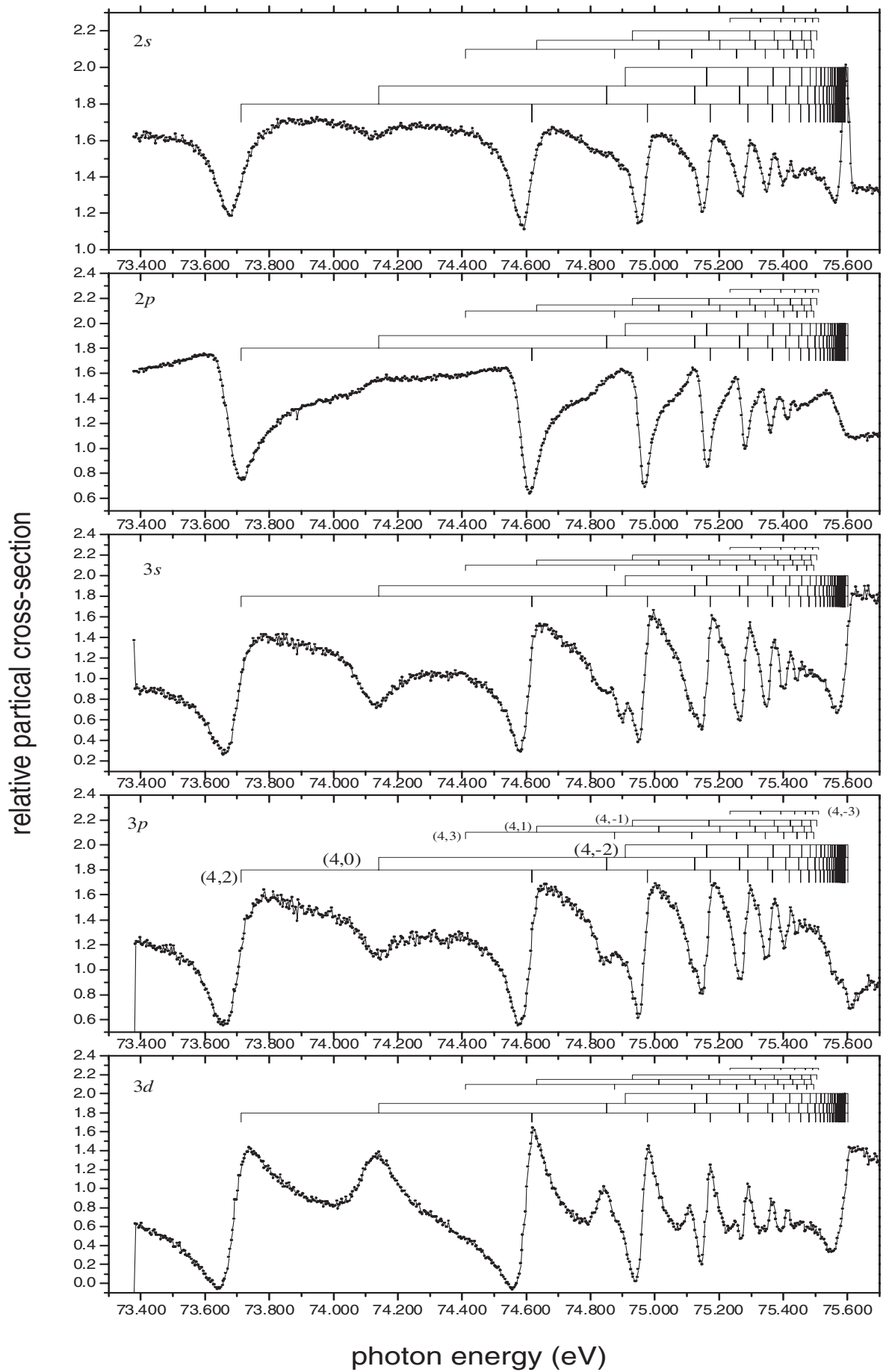


Figure 2  
Magnitudes of the 5 components separated as in Fig. 1 as a function of incident photon energy. The excitation energies of the doubly-excited states of interest are also shown.

of such a scan are shown in Fig. 2. These results confirm recent state-of-the-art theoretical calculations, and represent a first-step towards using time-resolved measurements to give a new dimension to studies of photo-ionisation and photo-excitation processes. Further details can be found in references 2 and 3.

**J. R. Harries<sup>1</sup>, J. P. Sullivan<sup>2</sup>, P. Hammond<sup>3</sup> and Y. Azuma<sup>4</sup>, (<sup>1</sup>JASRI, <sup>2</sup>Australian National Univ., <sup>3</sup>Univ. of Western Australia, <sup>4</sup>KEK-PF)**

## References

- [1] N. Terazawa, M. Kouchi, M. Ukai, K. Kameta and Y. Hatano, *J. Chem. Phys.*, **100** (1994) 7038.
- [2] J. R. Harries, J. P. Sullivan, S. Obara, P. Hammond and Y. Azuma, *J. Phys. B*, **36** (2003) L319.
- [3] J. R. Harries, J. P. Sullivan, S. Obara, Y. Azuma, J. G. Lambourne, F. Penent, R. I. Hall, P. Lablanquie, K. Bucar, M. Zitnik and P. Hammond, *J. Phys. B*, **38** (2005) L153.