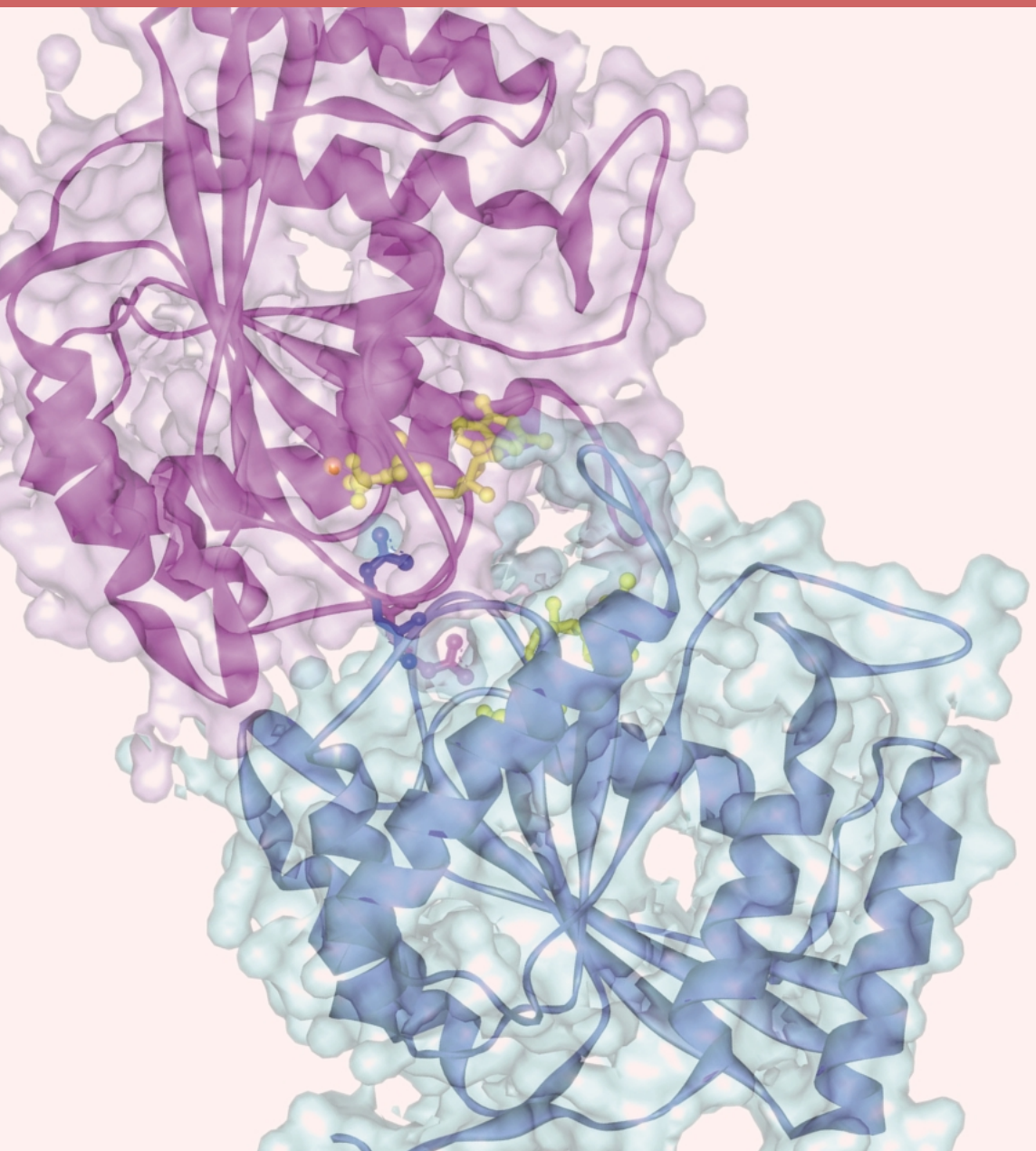
Highlights



1 Atomic and Molecular Science

1-1 The Stark Quantum Beat of Ne Fluorescence in the Vacuum Ultra Violet Region

In an electric field, energy levels of atom and molecule shift (Stark shift) and, degenerate energy levels split (Stark splitting). In the case of Ne, the ground state has total angular momentum J=0 and Rydberg states which is excited by photoexcitation of ground state have J=1. Both the ground and Rydberg states cause Stark shift. While Stark splitting occurs only in a excited Rydberg state because the J=1 state has the M=0 and $|M|$ =1 state as magnetic sublevels.

In the VUV region absorption measurements for Stark effect, resolution is now approximately 0.1 cm^{-1} (3 GHz) [1]. Although such absorption measurements are very high resolution spectroscopy, Stark splitting measurements require higher resolution spectroscopy. Therefore we used Stark quantum beat (SQB) spectroscopy in order to measure the Stark splitting. We can determine the tensor polarizabilities [2] and investigate the excited state decay dynamics with fluorescence in an electric field by applying the SQB technique. The quantum state controlling which is basic technique for quantum computing is one of the aims of our study.

Figure 1 represents the four-level system in the case of Ne atom. A short light pulse of appropriate frequency

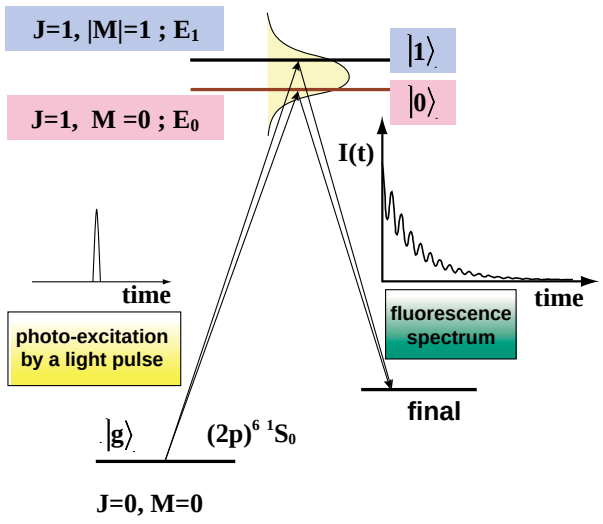


Figure 1
The |0> and |1> states are coherently excited from a single ground state |g>. The coherence is demonstrated by an interference effect (quantum beats) when photon decay to a common final state is observed.

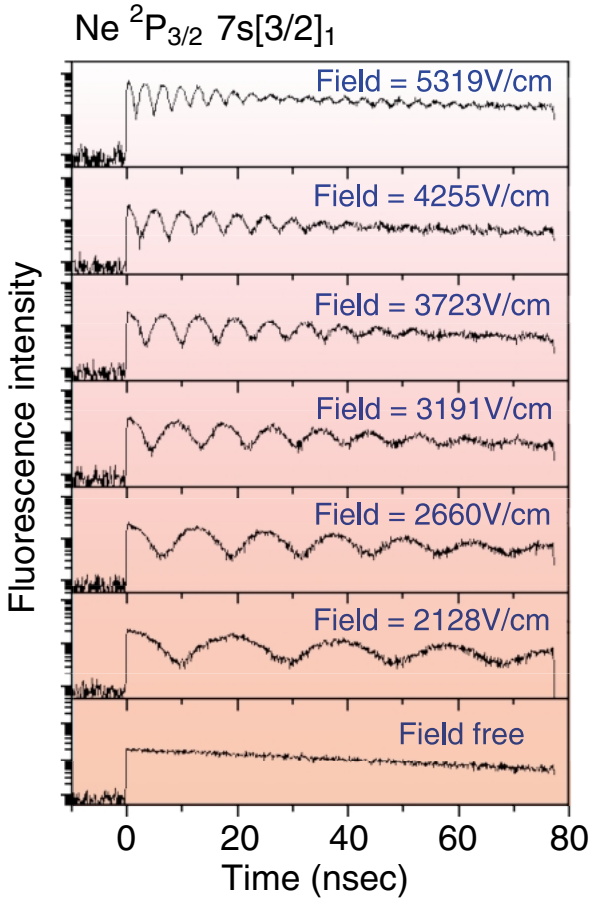


Figure 2
The fluorescence decay curve of the $7s[3/2]_1$ states obtained with polarized radiation slanted at +45° to the z axis and at different electric field strengths.

and of bandwidth larger than the energy splitting will coherently excite the two magnetic sublevels |0> and |1>. The SQB is caused by quantum interference effects in coherently prepared magnetic sublevels, and appears in fluorescence time spectrum.

The SQB measurement was performed on BL-20A by using PF ring single bunch operation. We used Stark plates which could rotate around the light axis and applied a static electric field up to about 5 kV/cm to the interaction region and used a MCP as the fluorescence detector [3]. Energy splitting of more than 10^{-4} cm^{-1} (10 MHz) between magnetic sublevels of Ne Rydberg states was measured in this study.

Figure 2 represents the fluorescence decay curves of the $7s[3/2]_1$ Rydberg state. This spectrum was obtained with polarization radiation angled approximately +45° to the z axis at several different field strengths. Since only fluorescence emitted from Rydberg states of total angular momentum J=1 were observed and Ne atom has no

hyperfine structure, the quantum beats recognized in the figure are caused by the quantum interference effects between the M=0 and $|M|$ =1 magnetic sublevels.

The Stark $|\Delta M|$ =1 beat fluorescence intensity is derived by the following equation [4],

$$I(t) \propto \varepsilon_x \varepsilon_z \lambda_x \lambda_z \cos(\omega t) \exp(-\Gamma t) \left| \langle J_e M_e \| \mu \| J_i M_i \rangle \right|^2 \left| \langle J_e M_e \| \mu \| J_f M_f \rangle \right|^2$$

where, $\langle J_e M_e |$, $|J_i M_i\rangle$ and $|J_f M_f\rangle$ represent the excited, initial, and final states, respectively. ω is the angular frequency corresponding to the energy split between the M=0 and $|M|$ =1 magnetic sublevels and Γ is the decay constant of the excited states. ε_x and ε_z are electric field amplitudes of excitation light along the x and z axes, respectively. The detection direction is characterized by cosines λ_x and λ_z which specify the observed fluorescence polarization directions. The factor λ_z has opposite signs for fluorescence emitted to one side and the other of the yz plane. That is, emitted fluorescence to opposing sides have opposite beat phases.

The 7s splitting field dependence is 11.04±0.23 MHz/(kV/cm)^2. This frequency at 1 kV/cm corresponds to the M=0 and $|M|$ =1 energy splittings of (368±7.7) $\times$ 10^{-6} cm^{-1}. A comparison was performed between the measured experimental energy splitting and our calculations applying the second order perturbation and using the *jl* coupling scheme and tables for the computation of radial integrals in the Coulomb approximation [5]. The calculated energy splitting of $7s[3/2]_1$ state is 10.985 MHz at 1 kV/cm electric field and agrees well with the experimental one.

The tensor polarizabilities can be deduced from our experimental value. The absolute value of $7s[3/2]_1$ Stark tensor polarizability was 7.36 MHz/(kV/cm)^2 with an uncertainty accuracy of ±4%.

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References

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1-2 Double Photo-Excitation of Helium in a Strong DC Electric Field

Photo-excitation of atoms in an electric field has been studied since the beginning of atomic spectroscopy as a sensitive test of our theoretical understanding of excited states. Nevertheless, previous studies all concerned single electron photo-processes, and the effect of an external electric field on highly correlated atomic processes remained completely unexplored.

The double photoexcitation resonances of helium have been well studied since the advent of synchrotron radiation as the prototypical atomic system dominated by electron correlation, providing an important test of our theoretical understanding of few body systems. Nevertheless, their evolution under an electric field has never been extensively studied experimentally. When compared to singly-excited Rydberg resonances, doubly-excited resonances have the additional complication that the competition between the electron-electron interaction and the applied field may be significant. In addition, the multitude of decay channels involving autoionization as well as the recently discovered fluorescence decay channels, which include those that lead to metastable He states, are all very strongly affected by the external electric field.

At the Photon Factory, we have initiated a research program on photoexcitation of atoms in an external electric field. A new apparatus (Figs. 3 and 4) was developed in collaboration with Dr. Peter Hammond at the University of Western Australia, and technical expertise regarding the handling of high-voltage was provided from the KEK Accelerator Laboratory. The apparatus employs a He gas beam crossing the photon beam at right angles. A voltage of up to 120 kV is applied to one of the two electrolyte-polished 120 mm diameter electrodes which is placed approximately one centimeter from an earthed electrode across the photon interaction region. The detection is accomplished through three channels. The earthed electrode is slotted for the efficient collection of photoions by a micro-channel plate (MCP) placed behind. In addition, new capabilities for fluorescence detection and metastables detection are incorporated, following the recent discovery of the significance of those decay channels [1]. A second MCP is placed downstream of the He beam and detects neutral particles, including photons and metastable atoms that travel in the direction of the atomic beam after the decay of the doubly excited

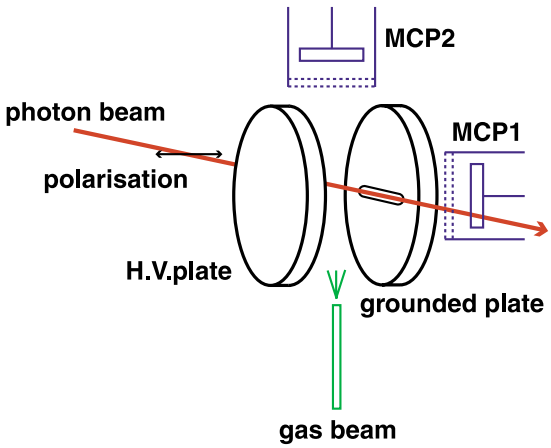

Figure 3
A schematic diagram of the apparatus.

Figure 4
A picture of the apparatus. During the experiment, the large high-voltage feed-through is covered by an aluminum box with copper-foil inner-lining to shield the rf-noise.

helium. The time-structure of the single-bunch operation of the synchrotron ring is utilized to efficiently separate fluorescence photons, which arrive with the speed of light and a small temporal spread forming a sharp peak in the time-of-flight (ToF) spectrum, and metastable atoms that travel much more slowly with significant temporal spread, forming a broad background.

The development of the apparatus and the experiments proceeded at BL-3B, BL-16B, BL-20A at the Photon Factory, and most recently at BL10.0.1 of the Advanced Light Source. We have measured the photo-ion yield spectra, fluorescence yield spectra and metastable yield spectra with different electric fields (Fig. 5). Pronounced electric field effects were observed in the metastable channel and fluorescence channel for the region close to threshold. With a moderate field of 10 kV/cm, the strength of the "b" ("–") series and "c" ("nd") series were drastically reduced, probably by enhanced autoionization due to the mixing with the main "a" ("+") series. The large threshold feature previously observed in the metastable channel [1] had disappeared.

Photoion spectra at varying field strengths were successfully measured in the region below the N=2 threshold and pronounced shifts of the resonance positions and the emergence of nominally forbidden (at zero field) resonances were observed for the first time (Fig. 6). The region between the 6a and 7a resonances was observed at field strengths of up to 84.4 kV/cm

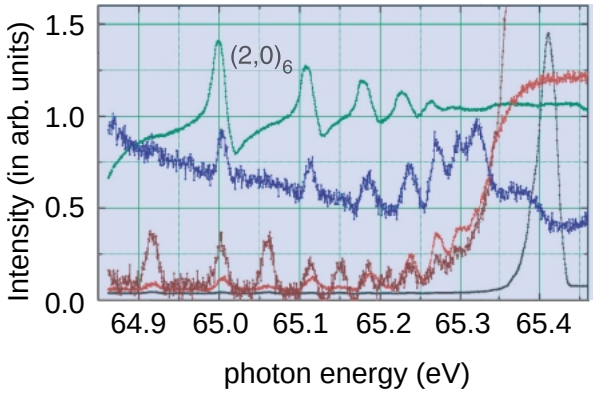

Figure 5
Ion (green), fluorescence (red) and metastable (blue) yield in a 10 kV/cm electric field. Compared to zero field metastable spectra (black and brown/expanded). The "–" and "nd" peaks clearly seen in the zero field metastables spectra are washed out with electric field.

and the evolution of the Stark splitting and mixing of the states in this energy region was mapped out as a function of field strength [2]. The behaviour of most of the predicted resonances agreed well with theory [3]. However, the emergence of a number of resonances not predicted by theory requires further investigation. Improvement in the theoretical treatment of this system is needed, in particular by considering the effect of the triplet resonances and the photon decay channel.

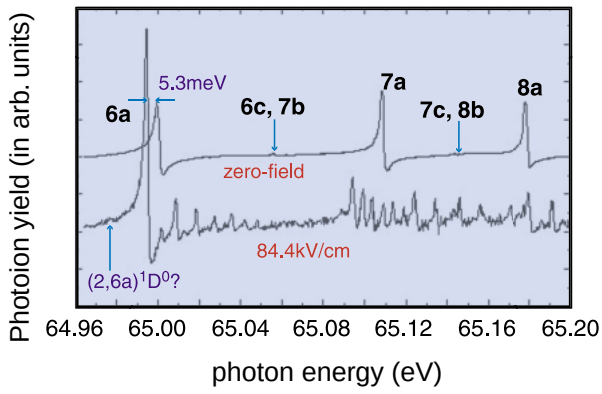

Figure 6
Photoion spectrum at zero-field and with an applied electric field of 84.4 kV/cm in the region of excitation to the 6a and 7a doubly excited states. Scales and offsets are chosen for maximum clarity.

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References

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