Exciton induced desorption of excited atoms in $2p^5 3p$ state from solid Ne

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INTRODUCTION

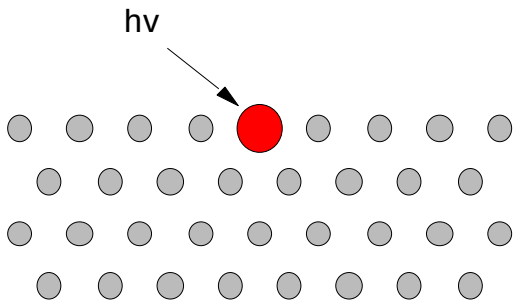
Desorption of excited atoms from the surface of rare gas solids by the creation of excitons has been extensively studied in these 10 years. Most of the experimental results have been well described in the framework of two proposed desorption mechanisms, cavity ejection (CE) and excimer dissociation (ED) [Figure 1].

Systematic measurements of TOF spectra of desorbed metastable Ne atoms (Ne* [$2p^5 3s\ ^3P_{0,2}$]) from the surface of solid Ne shows that the kinetic energy of desorbed atoms depends on the electronic excited states at desorption [Table 1, Figures 3-5]. At the excitation of $2p^5 3p$-type surface exciton (S'), the kinetic energy varies with the excitation energy [Figures 5, 6]. This shift of the peak position is likely due to the ten different energy levels in $2p^5 3p$ configuration [Figure 6, Table 2]. This result is in consistent with the cavity ejection scheme, i.e., the kinetic energy of desorbed atoms depends on the repulsive interaction between an excited atom and the surrounding ground state atoms.

Desorbed $2p^5 3p$ atoms are known to decay to $2p^5 3s$ states by emitting visible light in vacuum [Table 2]. Figure 8 shows a 3p -> 3s fluorescence spectrum of Ne* atoms in $2p^5 3p$ states desorbed from solid Ne by electron impact with the incident energy of 150eV, and by the selective excitation of S' excitons (λ = 64.2 nm) using monochromatic VUV light.

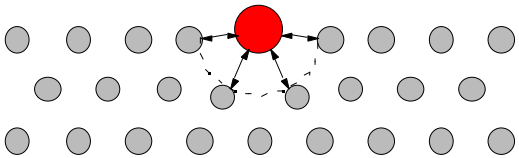
The purpose of the present study is State-Resolved Time-Of-Flight (SR-TOF) Spectroscopy, e.g., the measurements of TOF spectra of desorbed metastable atoms in coincidence with the 3p -> 3s visible fluorescence, from which we expect to get the detailed information on the correlation between the initial excitation and resulting desorption dynamics. Figure 9 shows the results for the first trial of the SR-TOF spectroscopy.

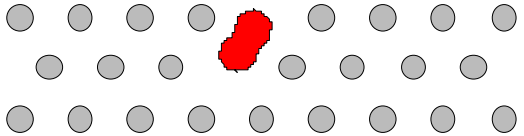
Cavity Ejection (CE) and Excimer Dissociation (ED) Mechanism



Exciton creation
on the surface

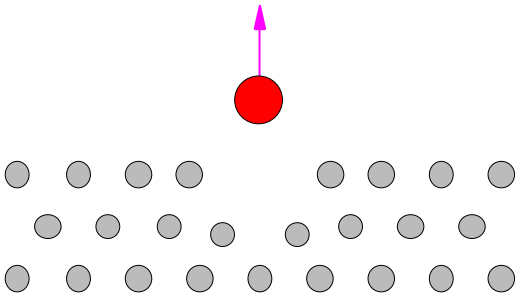
Cavity Formation

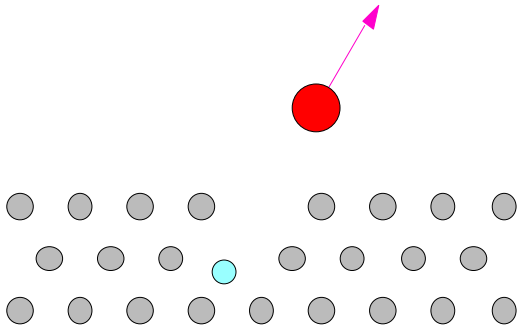




Excimer Formation

Desorption





Dissociation
&
Desorption

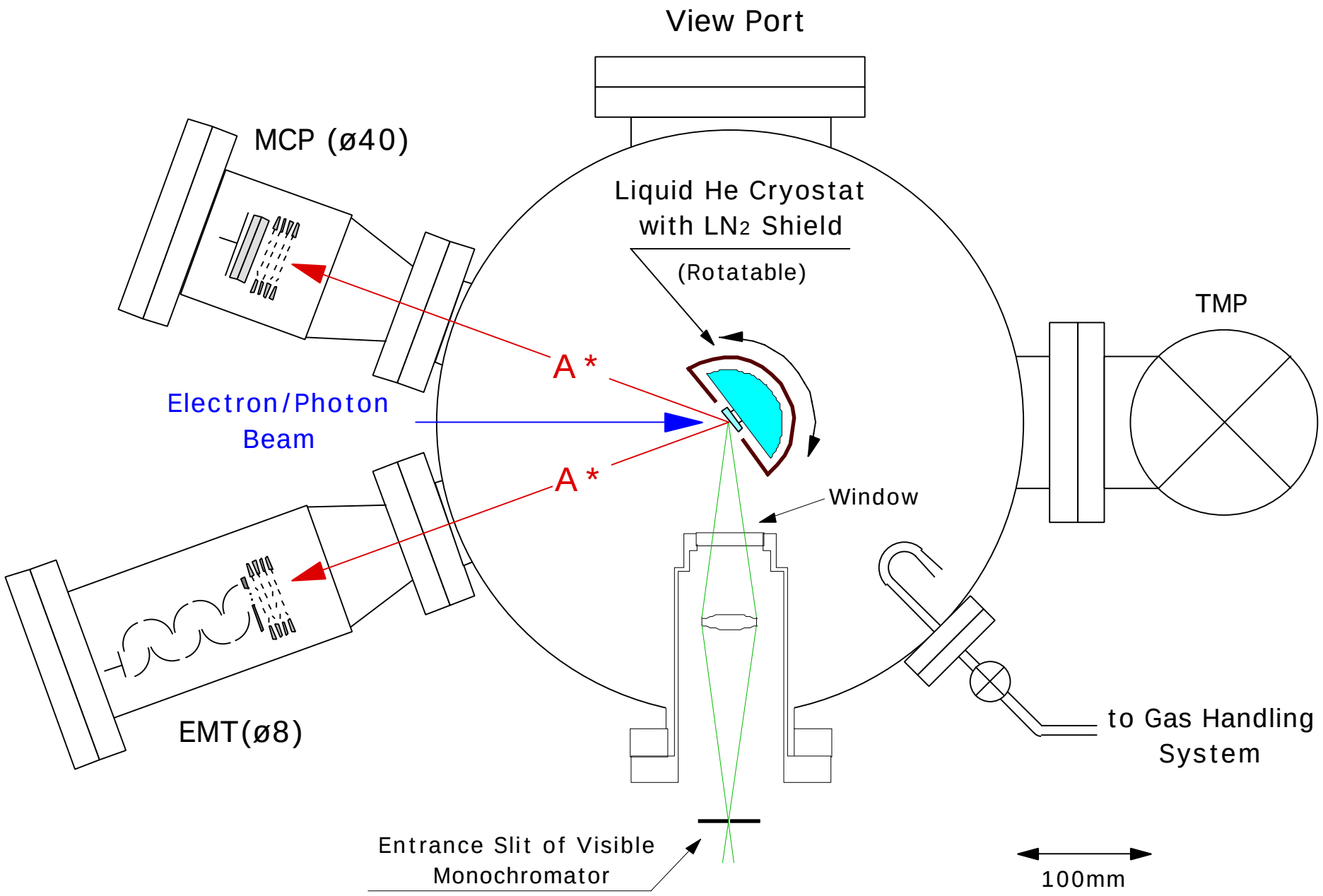
CE

ED

Kinetic Energies and Electronic States of Desorbed Ne*

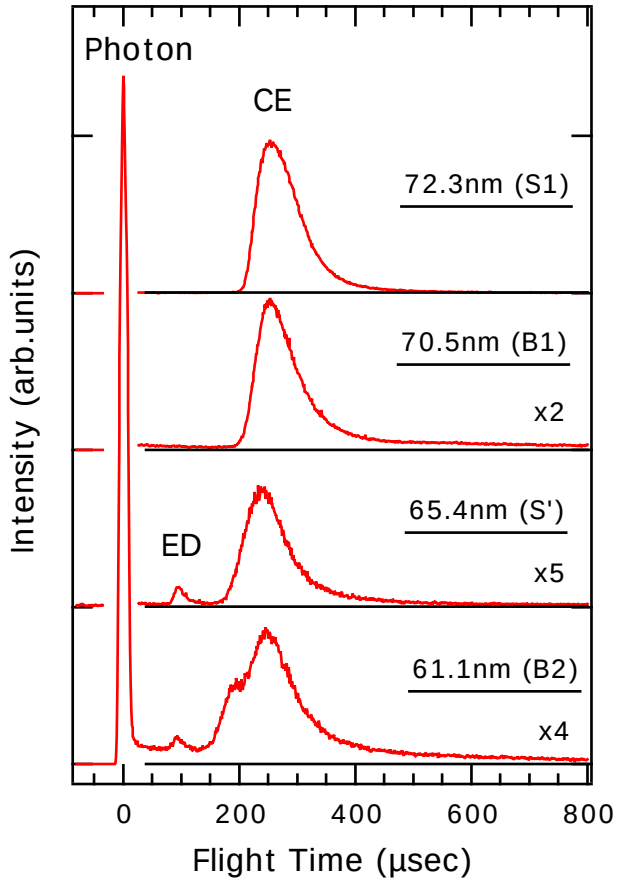
	Electronic Configuration	Excitation Energy	Kinetic Energy (eV)	Mechanism	Electronic State at Desorption (*tentative*)
S1	$2p^5 3s$	17.2 eV (72.3 nm)	0.18	CE	$2p^5 3s\ (^3P_{0,2})$
B1	$2p^5 3s$	17.5 eV (70.7nm)	0.18	CE	$2p^5 3s\ (^3P_{0,2})$
S'	$2p^5 3p$	19.0 eV (65.4nm)	0.19 0.2 ~ 0.4 1.5	CE (bulk?) CE ED	$2p^5 3p$ $2p^5 3p$
B2	$2p^5 4s$	20.3 eV (61.1nm)	0.18 0.36 1.5	CE CE (?) ED	$2p^5 3s\ (^3P_{0,2})$ $2p^5 4s$ (?)
B3	$2p^5 5s$	20.9 eV (59.2nm)	0.18 0.36 1.5	CE CE(?) ED	$2p^5 3s\ (^3P_{0,2})$ $2p^5 5s$(?)

Schematic Diagram of the Experimental Apparatus (Top View)

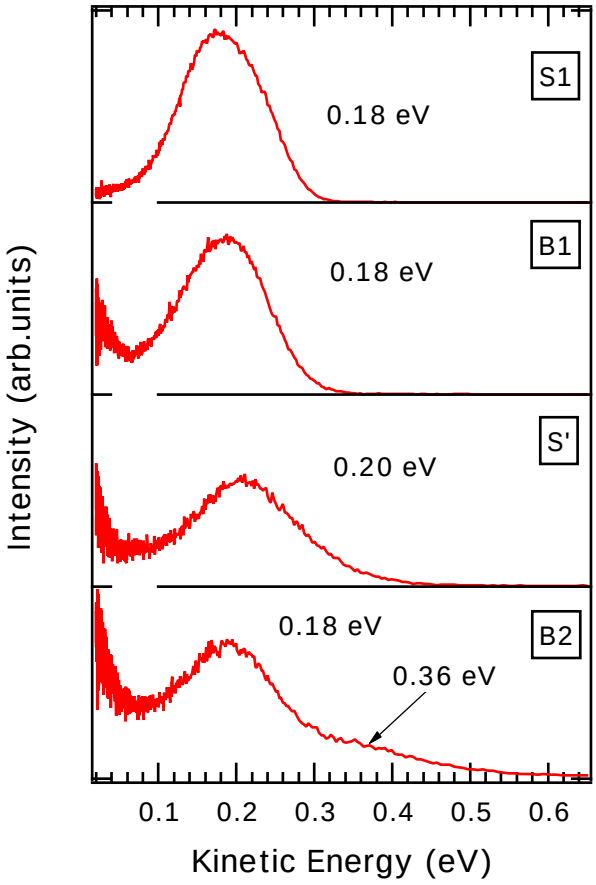


Photon Stimulated Desorption of Excited Atoms from solid Ne

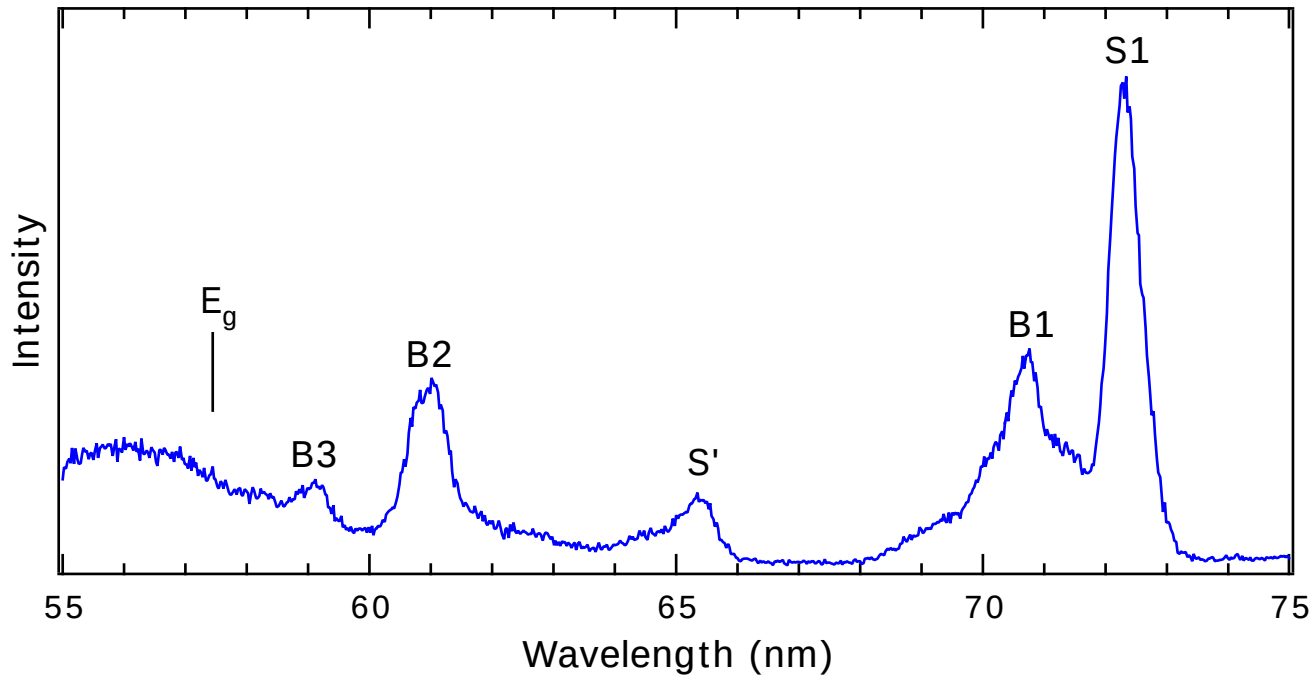
Time-of-flight Spectra



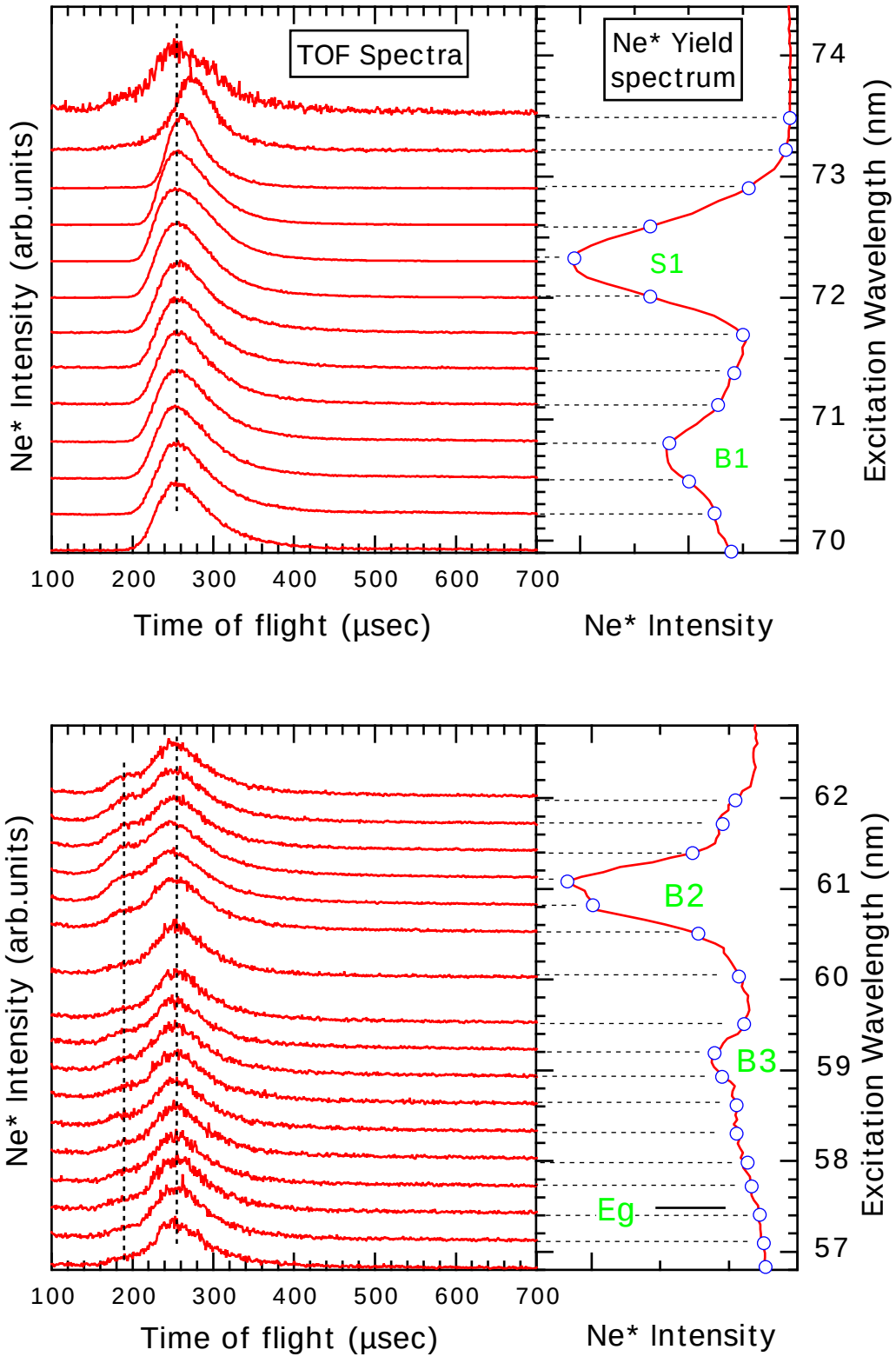
Kinetic Energy Spectra



Desorption yield of Ne*(CE) as a function of wavelength

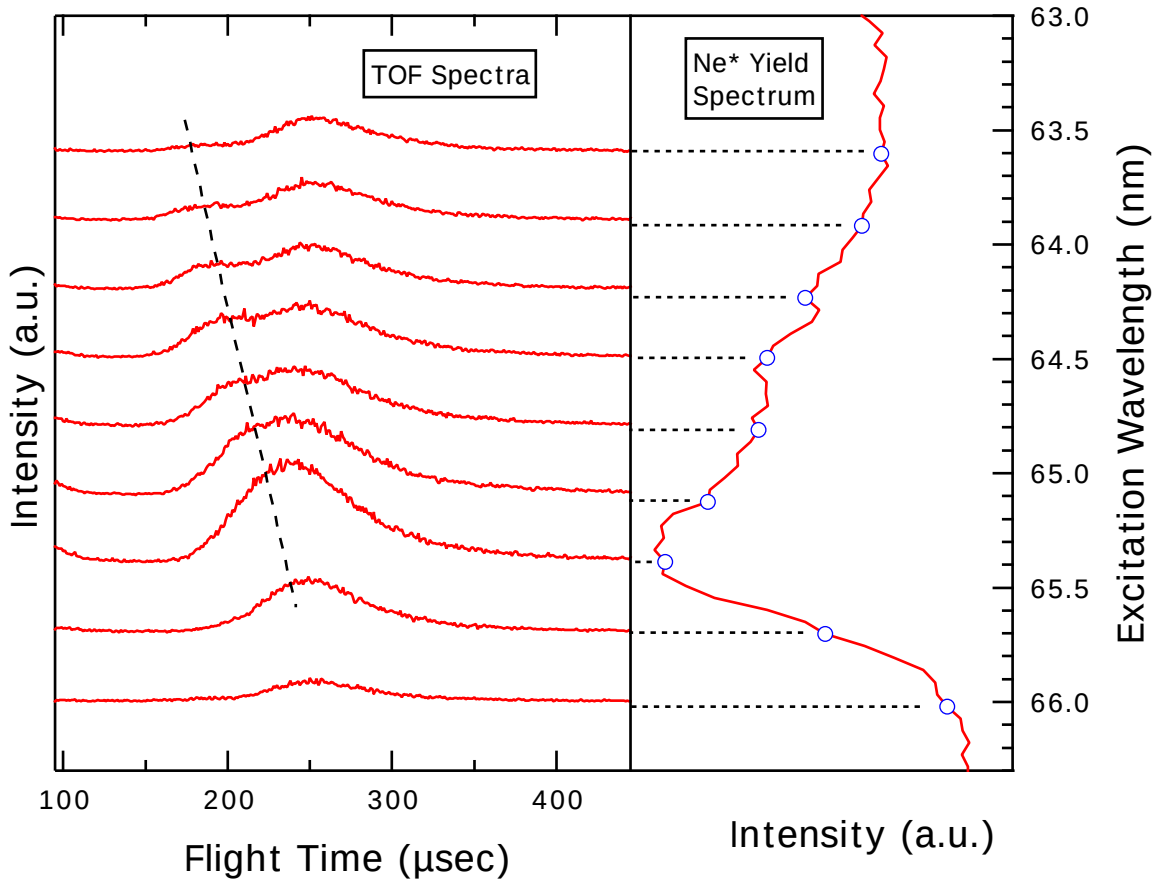


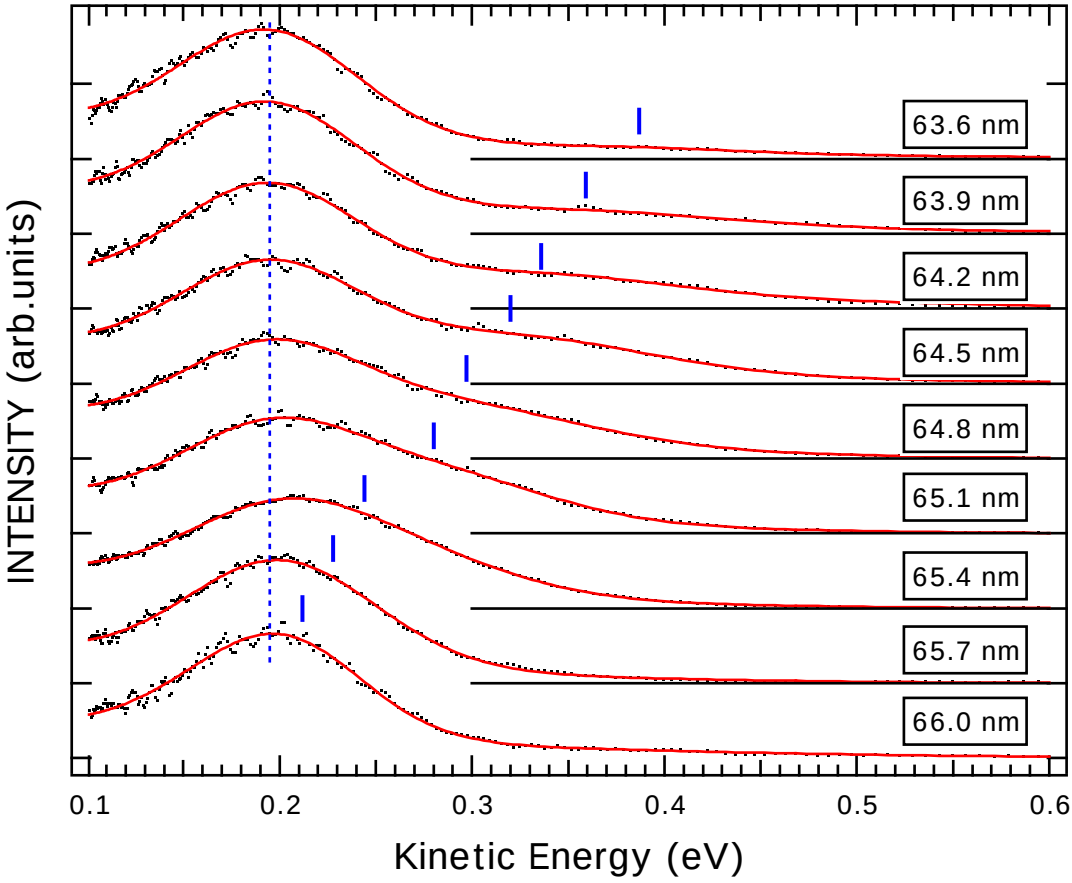
Series of Time-of-Flight Spectra of Desorbed Ne*(CE)



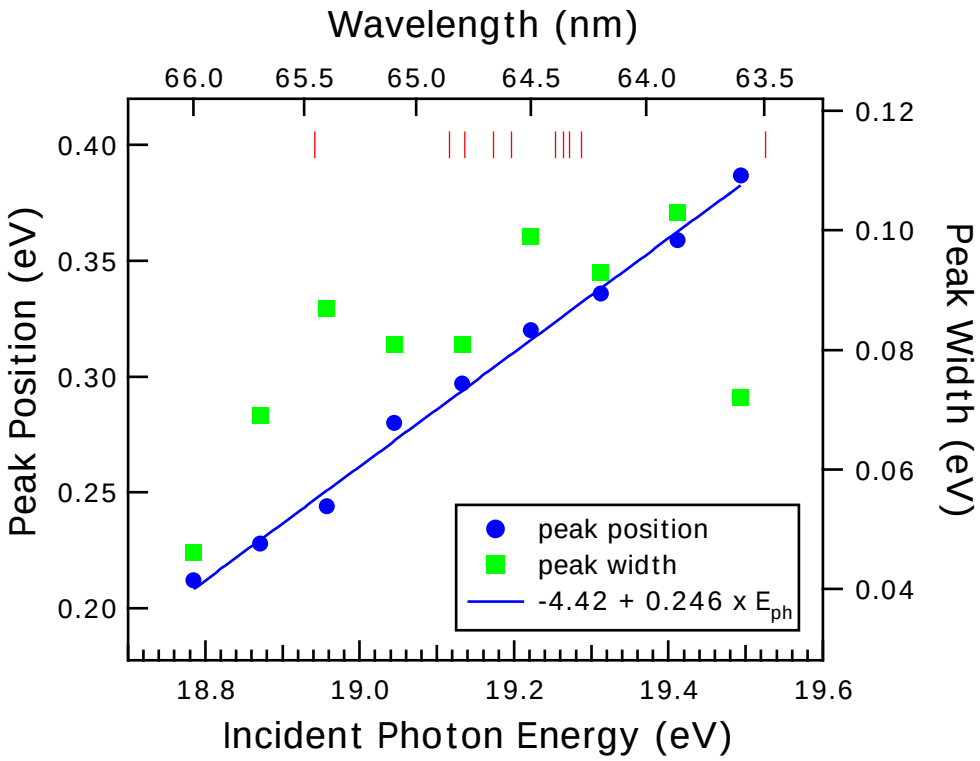
Series of Time-of-Flight spectra of desorbed Ne*(CE) by photon impact. Note that the peak position in TOF spectra does not depend on the excitation wavelength.

Series of Time-of-Flight and Kinetic Energy Spectra at the excitation of S' exciton.

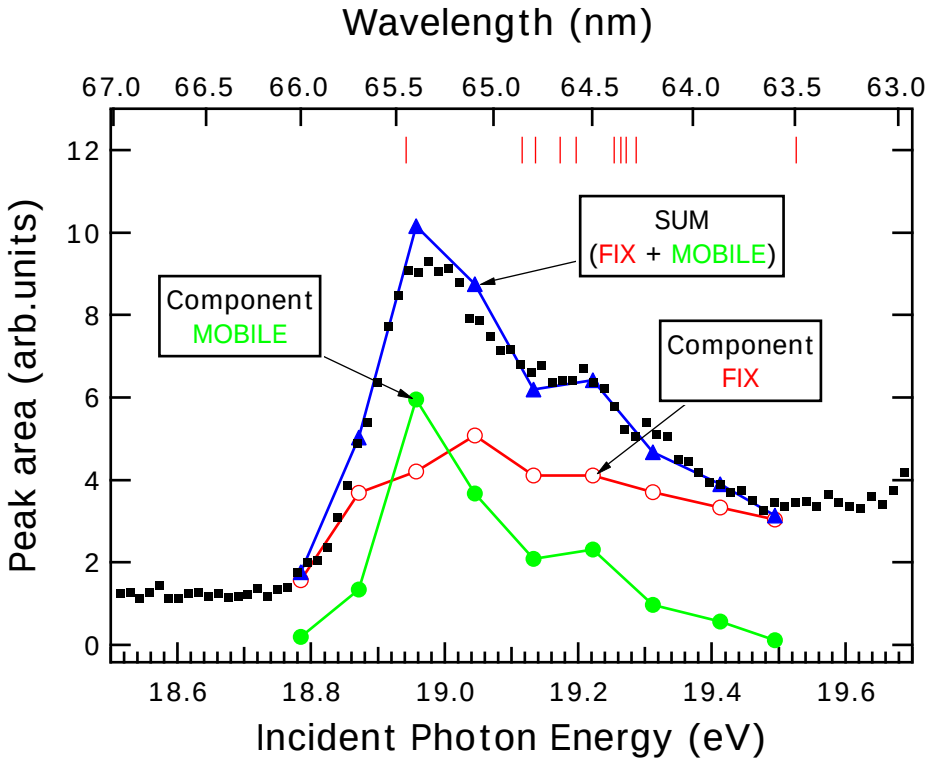




Position and width of the 'mobile' peak as a function of excitation wavelength.

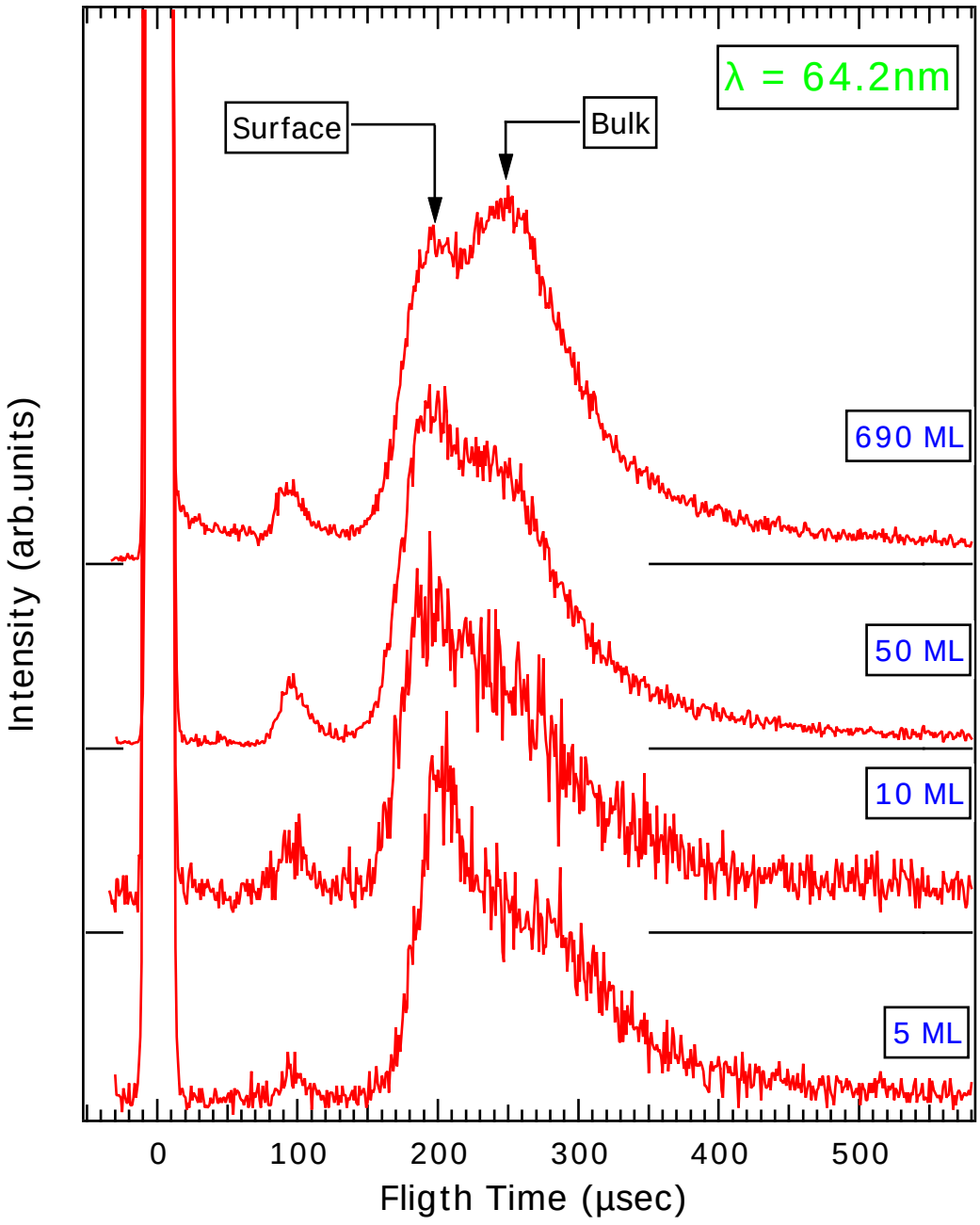


Peak area for each component as a function of excitation wavelength.



Dots represent the desorption yield of Ne*(CE) from Fig. 3.

Thickness dependence of TOF spectra of desorbed Ne*.



Thikness dependence of TOF spectra of Ne* desorbed from the surface of solid Ne. Faster and slower components show 'surface-like' and 'bulk-like' behaviour, respectively.

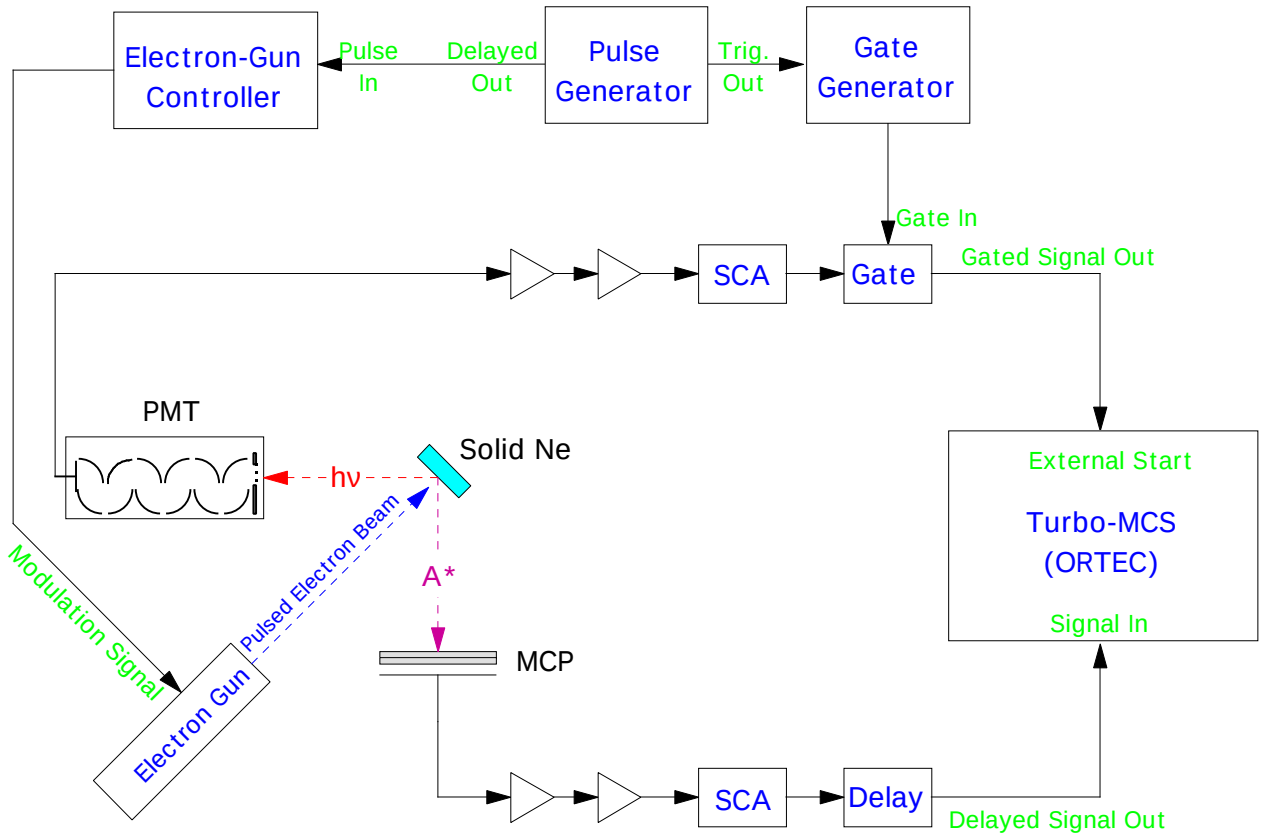
State-Resolved Time-of-Flight Spectroscopy [SR-TOF]

Detection of desorbed metastable atoms in $2p^5 3s\ (^3P_{0,2})$ state
[*Final State resolved*]

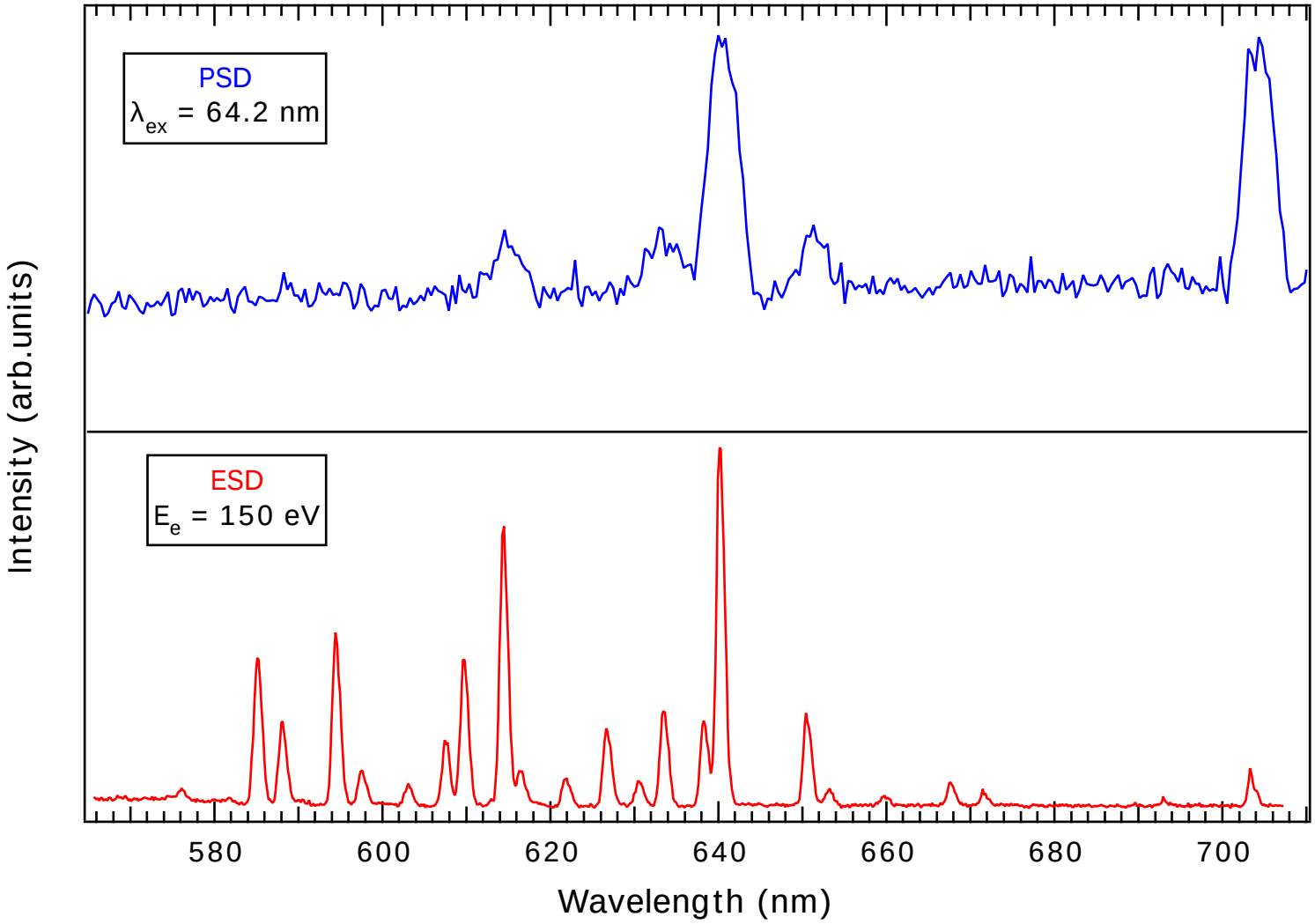
in coincidence with

3p -> 3s visible emission
[*Initial State resolved*]

Block Diagram of SR-TOF system in ESD mode



3p -> 3s fluorescence spectra by PSD (λ_{ex} = 64.2 nm) and ESD (Ee = 150 eV)



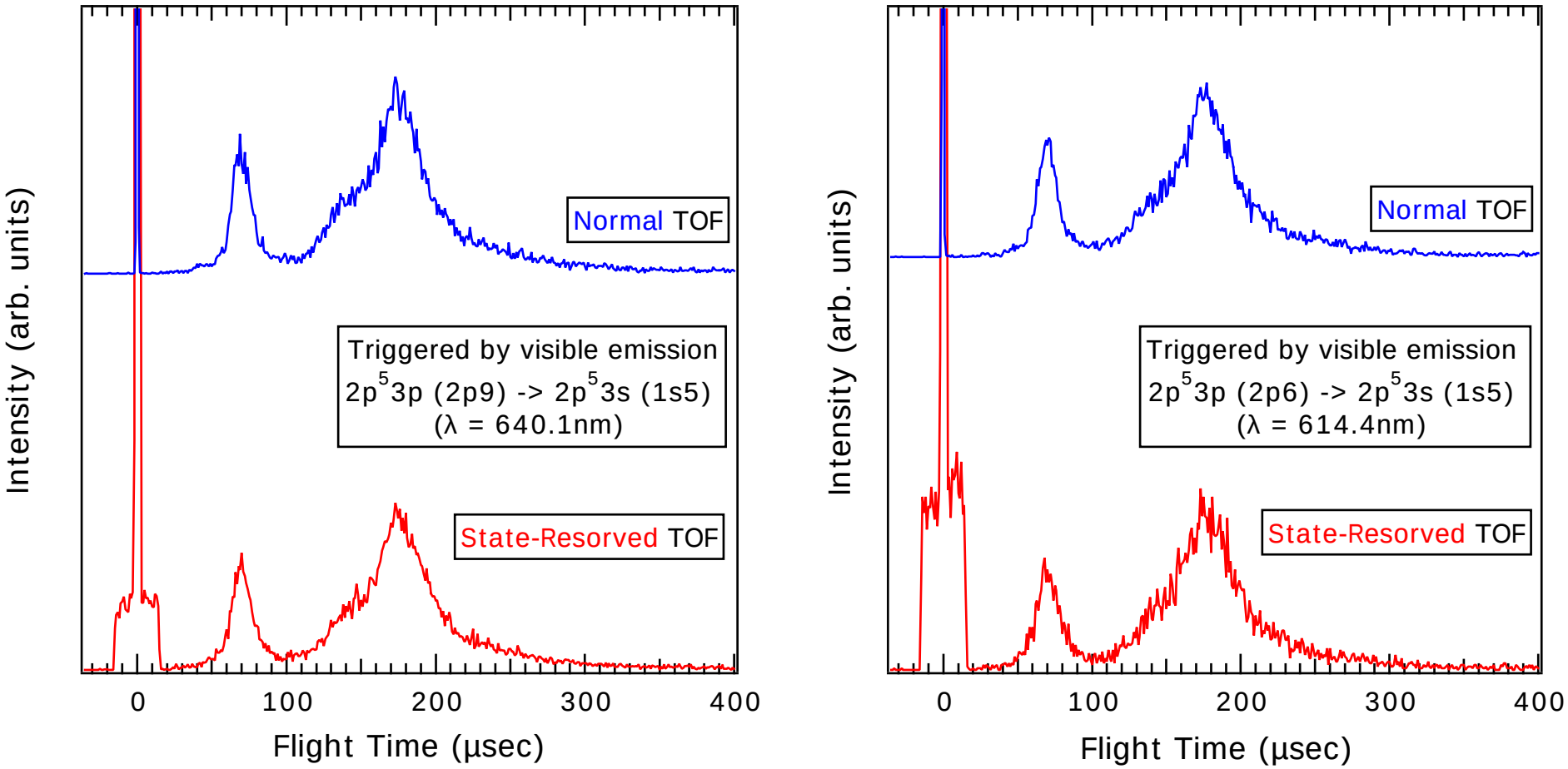
Excited states of Ne atom in the configuration of $2p^5 3s$ and $2p^5 3p$

Term (Paschen)	Energy (eV)	(nm)	Configuration	J	Lifetime (sec)	Transition	Wavelength of emission (nm)
1s5	16.619	74.60	$2p^5(^2P_{3/2})3s$	2	24.4		
1s4	16.671	74.37	$2p^5(^2P_{3/2})3s$	1	2.10×10^{-8}		
1s3	16.716	74.17	$2p^5(^2P_{1/2})3s$	0	430		
1s2	16.848	73.59	$2p^5(^2P_{1/2})3s$	1	1.51×10^{-9}		
2p10	18.382	67.45	$2p^5(^2P_{3/2})3p$	1	2.63×10^{-8}	1s2, 1s3, 1s4, 1s5	808.2, 744.2, 724.6, 703.2
2p9	18.556	66.81	$2p^5(^2P_{3/2})3p$	3	2.13×10^{-8}	1s5	640.1
2p8	18.576	66.74	$2p^5(^2P_{3/2})3p$	2	1.90×10^{-8}	1s2, 1s4, 1s5	717.5, 650.8, 633.5
2p7	18.613	66.61	$2p^5(^2P_{3/2})3p$	1	2.1×10^{-8}	1s2, 1s3, 1s4, 1s5	702.4, 653.6, 638.4, 621.8
2p6	18.637	66.52	$2p^5(^2P_{3/2})3p$	2	1.90×10^{-8}	1s2, 1s4, 1s5	693.0, 630.6, 614.4
2p5	18.694	66.32	$2p^5(^2P_{1/2})3p$	1	1.92×10^{-8}	1s2, 1s3, 1s4, 1s5	671.6, 626.8, 612.9, 597.5
2p4	18.704	66.29	$2p^5(^2P_{1/2})3p$	2	1.98×10^{-8}	1s2, 1s4, 1s5	668.0, 609.8, 594.6
2p3	18.712	66.26	$2p^5(^2P_{3/2})3p$	0	1.65×10^{-8}	1s2, 1s4	665.1, 607.4,
2p2	18.727	66.20	$2p^5(^2P_{1/2})3p$	1	1.98×10^{-8}	1s2, 1s3, 1s4, 1s5	659.8, 616.5, 603.0, 588.1
2p1	18.966	65.37	$2p^5(^2P_{1/2})3p$	0	1.45×10^{-8}	1s2, 1s4	585.4, 540.2

Green colored lines are the candidates for the coincidence measurements.

Fig. 9

First trial of State-Resolved TOF spectroscopy by
Electron Stimulated Desorption (incident energy : 150eV)



Summary

1. Desorption of an excited atoms by the creation of $2p^5 3p$-type surface exciton (S') has been systematically studied.

2. Two components of kinetic energy of desorbed Ne* at S' excitation have been observed. 'Mobile' peak was found to be surface-like, and 'fixed' component showed a bulk-like feature probably due to a defect in the bulk.

3. Kinetic energy of desorbed excited atoms at S' excitation ('Mobile' component) depends sensitively on excited states created, showing the validity of cavity ejection mechanism.

4. In order to get detailed information on the correlation between the initial excitation and resulting desorption dynamics, State-Resolved Time-of-Flight Spectroscopy (SR-TOF) was proposed and performed. Because of large background signal (mainly emission from the sample), SR-TOF spectra has not been obtained yet. Modification of the experimental setup is in progress.