VUV Photoionization of Superfluid Liquid Helium Droplets at the ALS

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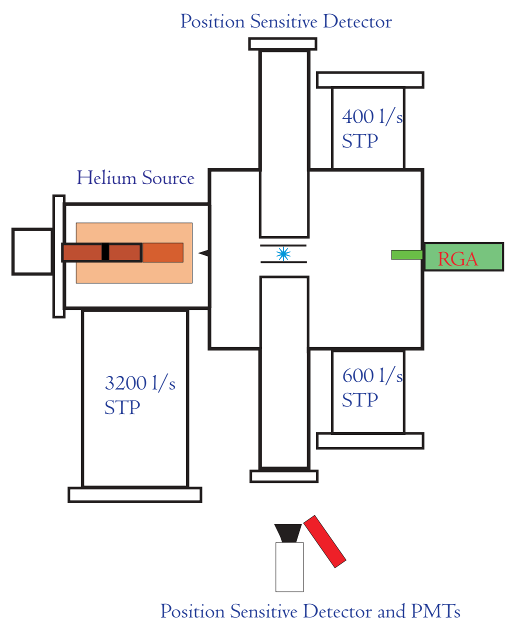
INTRODUCTION

Helium droplets have many interesting properties. They are very cold - evaporative cooling reduces the temperature to 380 milliKelvin. At these temperatures, the droplets are superfluid, and the electronic interactions between the He atoms can be described neither by He pair-potentials, nor by mean field theories. For an accurate description, quantum many body effects must be considered[1]. The complex interactions in the droplets will not affect only electronic excitations -relaxation processes will be altered as well. We have begun an in depth study of theses processes near the helium atomic ionization threshold.

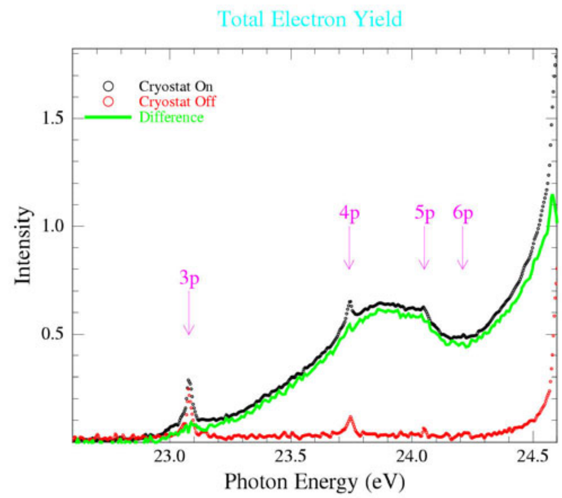
EXPERIMENTAL

The droplets were produced in a supersonic expansion using a 5 mm diameter nozzle aperture at typical He pressures of 15-50 bar and source temperatures of 11-18 K. The source conditions provided droplets with $\langle N \rangle = 10^4$ He atoms. The source chamber is pumped by a 3,200 l/s magnetically levitated turbo pump that is able to maintain a source pressure chamber of 10^{-4} with 60 bar, 13 K operation. The main chamber is evacuated by two magnetically suspended turbo pumps (Seiko Seike STP 600 and STP 400) backed by an oil-free scroll pump, providing 1000 l/s of pumping which keep the main chamber in the 10^{-7}'s during the experiment. VUV radiation from a 10 cm period undulator at the Advanced Light Source was dispersed by a 600 line/mm tungsten coated grating in a 3-m, normal incidence, off plane Eagle monochromator (McPherson). The endstation is schematically illustrated in the inset figure. The VUV light then ionized the droplets on the axis of a TOF spectrometer. The generated photoelectrons then struck a 40-mm-diameter dual multi-channel plate (MCP), which is coupled to a phosphor screen (Galileo 3040FM) yielding positional information. The electron optics were biased to achieve "Velocity Map" conditions[2]. A photomultiplier tube monitored the light from the phosphor screen, allowing for total ion yield measurements. The recorded images are 2-dimensional projections of the 3-dimensional recoiling product electron sphere. Axial symmetry is maintained around the polarization axis, allowing reconstruction of the images using conventional inverse Abel transform techniques. The droplet beam was also detected by a residual gas analyzer (SRS RGA 200), which was mounted in the main chamber along the droplet propagation axis.



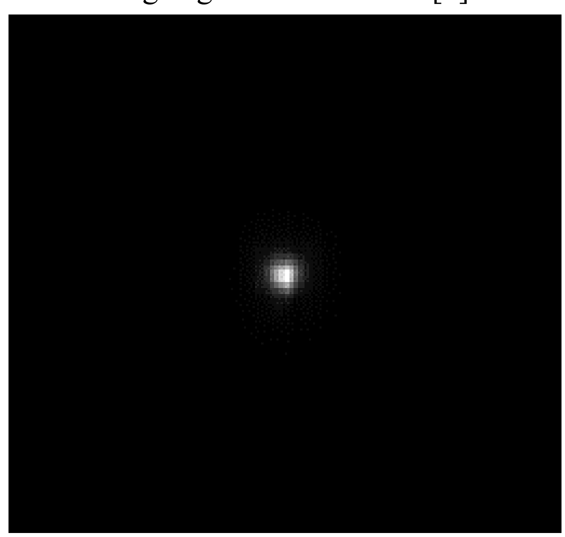
RESULTS and DISCUSSION

A photoelectron yield spectrum is shown to the right. The figure shows signal with the cryostat off - helium atoms only, the cryostat on - atoms and droplets, and the difference between the two. Also indicated in the figure are the atomic helium singlet *np* Rydberg energies converging the the helium ionization energy, 24.587 eV. Immediately one sees that the droplets ionization potential is suppressed relative to the atom.



The ionization process is mediated by the formation of He_2^+ which is bound by 2.35 eV relative to He + He^+. The photon energy required to access the minimum of the He_2^+ curve is ~22.2 eV. That the threshold for droplet ionization occurs ~.8eV above this is telling of the excitation process. In the droplets, the average distance between He atoms is ~3.6 Å. This is near the asymptotes of the He_2^* dimer potentials [3], thus it is expected that the excitations have significant atomic character. Using the atomic excitations as the zero order states of the droplets, we see the optically allowed excitation to He (3p) ^1P state is the first state of sufficient energy to allow for the production of He_2^+. Our ionization onset is slightly below that of the atomic He (3p) ^1P state (23.08 eV), this could result from a combination of factors. In the droplet, multibody effects can perturb both the energy and symmetry of the states and couple states of differing angular momentum. [4]

Examination of the photoelectrons lets us probe the photoionization process in more detail. The electron leaves the droplet with near zero kinetic energy (< 10 meV), with an isotropic angular distribution. The isotropic distribution contrasts with the dipole allowed excitation, which with linearly polarized light would have a cos^2 character. The electron angular and energy distributions change very little with with large changes in photon energy (23-24.5 eV). This implies that the excitation process is distinct from the ionization process, and is indirect.



Photoelectron image following droplet excitation at 24 eV . The angular distribution is isotropic, and the electrons have near zero kinetic energy (small image radius)

The initial excitation process generates essentially excited helium atoms in the droplets. These can relax via fluorescence, the dominant process [5], or they can feel the influence of neighboring atoms in the droplet, leading to the formation on He_2^*. Because the poor coupling of He_2^* vibrational modes to those of the droplets, significant energy remains in the He_2^*[6]. These states are energetically degenerate with $He_2^+ + e^-$. We believe this results in vibrational autoionization of the He_2*, and the subsequent generation of low kinetic energy electrons.

CONCLUSIONS

The first dispersed photoelectron spectra from the ionization of helium droplets have been collected, giving insight into the ionization process in liquid helium droplets. The isotropic, near zero energy electrons implicate an indirect ionization process. Ionization below the atomic He threshold results from autoionization of $He_2^*(He_n)$ states. Further work is underway to examine the relaxation of the vibrationally excited He_2^+ remaining in the droplet, and processes occurring above the He atom threshold.

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