ONE- AND TWO-PHOTON SPECTROSCOPY OF ALKALI ATOMS ON HELIUM NANODROPLETS AT 3 eV ENERGY

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We use the fundamental and second-harmonic of a ns-pulsed $\mathrm{Ti{:}Al_2O_3}$ laser with a 5 kHz repetition rate to measure the laser-induced fluorescence spectra of Rb atoms on the surface of superfluid helium nanodroplets.
Because of the shift and broadening induced by the droplet (the first in particular becoming increasingly large for these high-energy levels) assignment based on proximity to known gas-phase transitions is no longer reliable. The $5D$ and $7S$ levels in particular, both accessible by a two-photon transition, are sufficiently close in energy that even a description in terms of pure atomic states may be questionable.
We obtain supplementary information from gated-photon counting measurements of the emitted fluorescence. Because atoms dissociate from the droplet upon excitation, emission occurs from the gas phase, so we use the known gas-phase lifetimes of all levels involved to interpret our data.
We assign one band observed with the second-harmonic beam to the spectrum of the $6P \leftarrow 5S$ one-photon transition at ~ 24000 cm^{-1}. A second band observed with the fundamental beam at $\sim 2 \times 13000$ cm^{-1} is assigned to the $5D \leftarrow 5S$ two-photon transitions based on the measured lifetime of the cascading fluorescence. We further support the assignment with model calculations of the Rb-droplet interaction potential, taking the mixing of atomic states into account.