SPECTROSCOPY OF Li ATOMS AND Li DIMERS IN THE TRIPLET MANIFOLD ON THE SURFACE OF HELIUM NANODROPLETS

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Helium nanodroplets (He_N) have attracted strong interest as superfluid nanocryostats and can serve as a tool for the efficient preparation of tailored molecules and clusters[a]. Alkali-metal atoms and molecules are bound only weakly to the He_N surface. The fragility of these systems leads preferably to the formation of high-spin molecules on He_N[bc]. We use this property of helium nanodroplets for the preparation of Li dimers in their triplet ground state ($1^3\Sigma_u^+$). We present an excitation spectrum of the $2^3\Pi_g(\nu' = 0 - 10) \leftarrow 1^3\Sigma_u^+(\nu'' = 0)$ transition. The interaction between the molecule and the droplet manifests in a broadening of the transitions with a characteristic asymmetric form. The broadening extents to the blue side of each vibronic level, which is caused by the simultaneous excitation of the molecule and vibrations of the droplet (phonons). The two isotopes of Li form $^6Li_2$, $^7Li_2$ as well as the isotope mixed $^6Li^7Li$ molecule on the droplet surface. By using resonance enhanced multi-photon ionization time-of-flight (REMPI-TOF) spectroscopy isotope dependent effects could be studied.

In addition to excitation spectra of Li molecules, we report on the $3p \leftarrow 2s$ and $3d \leftarrow 2s$ two-photon transitions in single isolated Li atoms on He_N. From the 2S(Σ) ground state, two-photon transitions into Δ, Π and Σ molecular sub-states are possible. Mass dependent excitation spectra are recorded by using REMPI-TOF spectroscopy, which allows an investigation of the exciplex (Li*-He_m, m = 1-3) formation process in the Li-He_N system. Electronic states are shifted and broadened with respect to free atom transitions, which is explained within the pseudo-diatomic model.

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