Real-Time Autodetachment Dynamics of Vibrational Feshbach Resonances in Multipole-Bound States

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Abstract

We investigated vibrational Feshbach resonances of multipole-bound states in the time-domain for the first time. State-specific autodetachment rate measurement in phenoxide (PhO-) dipole-bound state (DBS) [1,2] and 4-cyanophenoxide (4-CP-) quadrupole-bound state (QBS) [3] was performed in the cryogenically-cooled ion trap combined with the time-resolved photoelectron velocity map imaging technique. For the phenoxide DBS, the most prominent $11'1$ ($+519 \text{ cm}^{-1}$) peak showed $\tau \sim 33.5$ ps. The lifetime of the each vibrational peak is highly mode-dependent to give $\sim 5$ ps for the $18'1$ ($+632 \text{ cm}^{-1}$), and the multi-quanta excitation of the $v_{11}$ mode gives $\sim 11.9$ ps for the $11'2$ ($+1036 \text{ cm}^{-1}$) and $\sim 9.2$ ps for the $11'3$ ($+1556 \text{ cm}^{-1}$). These mode-specific autodetachment rates are well-described by the Fermi’s golden rule. Interestingly, the $11'18'1$ combination mode showed bifurcation dynamics to the $110'181$ and $111'180$ neutral product with much faster autodetachment rate of $\sim 1.4$ ps. This result implies that the each bifurcated channel showed accelerated autodetachment dynamics compared to the detachment from the single-mode respectively.

For the 4-CP- QBS, the lifetime of the most prominent $12'1$ peak gives $\tau \sim 56.4$ ps lifetime, which is $\sim 1.68$-fold slower than the similar $11'1$ mode of phenoxide DBS. This may originate from the weaker interaction of the charge-quadrupole potential in long-range region compared to that of the charge-dipole interaction. The mode-dependency is also confirmed by measuring the lifetime on the other vibrational modes of QBS, which could be described by the wobbling of the quadrupole moment ellipsoid and Fermi’s golden rule. This research provides unprecedental insights into the interaction between the electronic and nuclear dynamics of the non-valence bound states.

Keywords: Feshbach resonance, autodetachment, non-valence bound state, Time-resolved photoelectron imaging

References: