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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 cm⁻¹) peak showed $\tau \sim 33.5$ ps. The lifetime of the each vibrational peak is highly mode-dependent to give ~ 5 ps for the 18⁻¹ (+632 cm⁻¹), and the multi-quanta excitation of the ν_{11} mode gives ~ 11.9 ps for the 11⁻² (+1036 cm⁻¹) and ~ 9.2 ps for the 11⁻³ (+1556 cm⁻¹). These mode-specific autodetachment rates are well-described by the Fermi's golden rule. Interestingly, the 11⁻¹18⁻¹ combination mode showed bifurcation dynamics to the 11⁰18¹ and 11¹18⁰ neutral product with much faster autodetachment rate of $\tau \sim 1.4$ 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 ~ 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 unprecedented 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

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