論文の内容の要旨

論文題目　All-optical selective formation of ultracold molecules in the rovibrational ground state
(振動・回転基底状態にある極低温分子の全光学的・選択的生成)

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This thesis describes the experimental realization of a novel approach for producing ultracold molecules in the rovibrational ground state. Photoassociation followed by stimulated Raman adiabatic passage (STIRAP) enabled us to achieve ultracold $^{41}K^{87}Rb$ molecules in the rovibrational ground state within 100 ms, which was faster by more than two orders of magnitude than previous methods based on magnetoassociation. The presented all-optical method opened up a new avenue for manipulating a wide variety of molecular species including diamagnetic atoms, which cannot be manipulated via previous methods.

We first describe the experimental procedure for producing and detecting ultracold weakly bound $^{41}K^{87}Rb$ molecules via the photoassociation of laser-cooled ^{41}K and ^{87}Rb atoms. The apparatus consisted of a vacuum chamber, optical setups for laser cooling, photoassociation lasers, a pulse laser for ionization, and ion-detecting devices. Combined with time-of-flight mass spectrometry, resonance enhanced multi-photon ionization (REMPI) enabled us to detect KRb molecules in a specific vibrational level.

The crucial step towards realizing ultracold molecules in the rovibrational ground state was an appropriated choice of the intermediate state for STIRAP. We found 10 vibrational levels (v''=41–50) of the $(3)^1\Sigma^+$ state via depletion spectroscopy for weakly bound molecules. A detailed analysis enabled us to construct an accurate potential energy curve of the $(3)^1\Sigma^+$ state as well as to identify the symmetry of the observed state as $(3)^1\Sigma^+$. Another candidate for the intermediate state, the $(2)^3\Sigma^+$ state, was also studied for 17 vibrational levels (v''=39–55). In each vibrational level, we observed structures of the order of 1 GHz, which were understood as rotational structures. In addition, we found that hyperfine structures due to the nuclear spin of ^{87}Rb played an important role in the levels with a low rotational quantum number (J' < 3).

Based on the spectroscopic studies, we developed a stable light source for STIRAP at two wavelengths (900 nm and 650 nm) which could connect weakly bound molecules and the rovibrational ground state. Diode lasers referenced to an ultralow expansion (ULE) cavity served as master lasers, which had a short-term linewidth below 10 Hz and a long-term stability better than 20 kHz. Other lasers, a diode laser

and a dye laser, were then locked to master lasers and were utilized for STIRAP experiments. By changing the offset frequencies between the master lasers and the slave lasers, we could scan the slave lasers over a few GHz with keeping the stability of the master lasers. The tunability and the stability realized in this setup were one of the key elements for realizing the STIRAP transfer of photoassociated molecules.

The most important issue for reaching the rovibrational ground state was a new spectroscopic method, which enabled us to precisely determine the transition frequencies from photoassociated molecules to the excited state $(3)^1\Sigma^+$. Further, this new method enabled us to observe the rovibrational ground state via two-photon dark resonance spectroscopy, which played a crucial role for determining the transition frequency as well as the transition strength between the excited state and the rovibrational ground state. Finally we demonstrated the selective formation of ultracold molecules in the rovibrational ground state via a STIRAP transfer with an efficiency of more than 70 %. The produced molecules were directly detected via REMPI ionization. The STIRAP transfer into the rovibrational ground state opened a new possibility for precisely measuring the hyperfine structures of weakly bound molecules in unexplored region with an accuracy of 10 kHz.