PRECISE MEASUREMENT OF THE VIBRATIONAL TRANSITION
FREQUENCIES OF HOMONUCLEAR DIATOMIC MOLECULAR IONS

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The precise measurement of molecular vibrational-rotational transition frequencies is useful for the development of new fields of physics like (1) search for the variation in the proton-to-electron mass ratio, (2) search of the electron electric dipole moment, and (3) search of the gravitational effect in the micro size etc.. However, precise measurement of molecular transition frequencies is much more difficult than atomic transitions. This is because the molecular vibrational-rotational energy structure makes it difficult to localize molecules in a selected quantum states and to reduce the kinetic energy by laser cooling.

In this paper, we show that the vibrational transition frequencies of homonuclear diatomic molecular ions (N$_2^+$, O$_2^+$) can be measured with the uncertainty lower than $10^{-17}$. The molecular ions can be prepared in a selected vibrational-rotational state by the Resonance Enhanced Multi-Photon Ionization. The kinetic energy is reduced by sympathetic cooling with laser cooled atomic ions. For the homonuclear diatomic molecular ion, the Stark shift is induced only by the coupling with electronic excited states, which is more than four orders smaller than that for the hetero-nuclear molecules.

The N$_2^+$ ($I = 0$) $^2\Sigma$ $(v,N,J,M) = (0,0,1/2, \pm 1/2) \rightarrow (v',0,1/2, \pm 1/2)$ ($v' = 1, 2, ...$) transition can be measured with the uncertainty lower than $10^{-17}$, because the Zeeman and electric quadrupole shifts are zero with a single transition. Here, $v$ and $N$ are the quantum number of vibrational and rotational states, respectively. $J$ is the total angular momentum including that of the electron. $M$ is the component of $J$ parallel to the magnetic field. $I$ is the nuclear spin, which can be 0 or 2 with even rotational state and 1 with odd rotational state (with $^{15}$N$_2^+$, $I = 2$ does not exist).

For the $^{16}$O$_2^+$ $^2\Pi_{1/2}$ $(v,J,M) = (0,1/2, \pm 1/2) \rightarrow (v',1/2, \pm 1/2)$ transition, electric quadrupole shift is zero and the Zeeman shift is of the order of $\pm 0.14 \times v'$ Hz/G. Averaging the $M = \pm 1/2 \rightarrow \pm 1/2$ transition frequencies, also this transition can be measured with the uncertainty lower than $10^{-17}$.

Measurement of these transition frequencies is useful for search of the variation in the proton-to-electron mass ratio.

References