SUBMILLIMETER-WAVE SPECTROSCOPY OF MOLECULAR IONS AND FREE RADICALS

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There is a long history of development of submillimeter-wave radiation sources such as frequency multiplication of lower frequency microwave or difference frequency generation by mixing either mid-infrared or far-infrared laser radiation with microwave. However, submillimeter-wave region is still relatively unexplored. The output power and frequency tunability have been major difficulties in routine application of the technique to high sensitivity spectroscopy. We at Ibaraki use backward-wave oscillators, phase-locked to stabilized millimeter-wave sources by means of GaAs harmonic mixers developed by Winnewisser group at Köln; the radiation sources deliver relatively high power of the order of mW and wide frequency tunability, making the system a powerful high-resolution and high-sensitivity spectrometer.

By combining various discharge sources such as a hollow cathode and an extended negative glow discharge cell with double modulation technique, we investigated submillimeter-wave spectra of free radicals such as CH, TiCℓ, HCS, FCO and NCS and ions like $C_2H_3^+$, H_2COH^+ and D_2H^+ in the past several years. Among these species, D_2H^+ is particularly interesting. Although the astrochemical significance of H_3^+ and its singly deuterated species, H_2D^+, is widely recognized and extensively investigated, spectroscopic information on the doubly deuterated species, D_2H^+, has not been as extensive. This ion is an important multiply deuterated species itself and at the same time it may play a crucial role in the chemistry of multiple deuteration. The first laboratory detection of a submillimeter-wave line of D_2H^+ will be discussed in terms of the recent discoveries of multiply deuterated molecules in extreme cold molecular clouds.