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Difference in cooling dynamics between photo-excited C_6^- and C_6H^-

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Synopsis Laser-induced electron detachment of C_6^- and C_6H^- stored in an ion storage ring was observed respectively, focusing attention onto the delayed processes with the time constants up to several milliseconds. We found the drastic difference in the decay profiles of photo-excited C_6^- and C_6H^- that is, C_6^- decayed much faster than C_6H^-. The unexpected fast decay of C_6^- indicates that it is governed by the radiative cooling with electronic transitions rather than vibrational nor depletion cooling.

Radiative cooling of molecules is a ubiquitous phenomenon in collision-free environment, for example in space, and is an indispensable process to complete the photon-excitation-relaxation cycle. Particularly, for carbon cluster anions and derivatives, detection of $C_{2n}H^-$ (n=2-4) in space has stimulated a wide interest in the radiative cooling of such species [1]. In the present study, we have measured the delayed electron detachment of photo-excited C_6^- and C_6H^-, to investigate their cooling processes.

The experiments were performed using an electrostatic ion storage ring at Tokyo Metropolitan University (TMU E-ring) [2]. Hot molecular anions of C_6^- and C_6H^- (energy: 20 keV) generated in a cesium sputter source were stored in the ring. After a specific storage time, the third harmonics of the Nd:YAG laser (355 nm) was merged at the one of the straight section of the ring, and at the same side, generated neutral particles were detected by a micro-channel plate.

The decay profiles of photo-excited C_6H^- and $C_6^-$are shown in Fig.1(a) and (b), respectively. The laser firing time is 6.5, 40 and 90 ms for each anion. There is a striking difference between the C_6^- and C_6H^-. The decay of C_6H^- is in the time range of 1 ms and shows the storage time dependence; the longer storage leads to slower decay. The rate of C_6H^- decay is consistent with the theoretical calculation based on the vibrational radiative cooling. In contrast, the decay of C_6^- is much faster than C_6H^-, in the time range of 10 μs, and it is not sensitive to the storage time. It is by far faster than theoretical prediction if the vibrational radiative cooling is solely taken into account. The different decay profiles between the species with similar heat capacities and detachment thresholds are most likely due to the difference in the importance of the electronic radiative cooling (recurrent fluorescence [3, 4]). A simple calculation based on the statistical weights of the excited states and the electronic transition probabilities indicates that, for C_6H^-, the contribution of the recurrent fluorescence is negligible since there is no low-lying excited state. For C_6^-, which has the open-shell electronic structure in its ground state, the low-lying electronic excited states highly enhance the cooling rate, making the calculated decay to well reproduce the experiments.

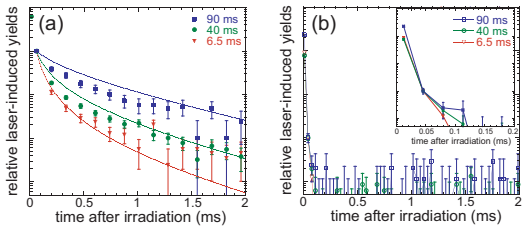

Figure 1. Storage time dependence of the decay profiles for (a) C_6H^- and (b) C_6^- enhanced by 355 nm laser irradiation. Simulated decay profiles of C_6H^- are given in (a) assuming that ions are cooled by vibrational transitions.

References

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