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Observation of recurrent fluorescence photons from small carbon cluster anions

Yuta Ebara*, Takeshi Furukawa*a 1, Jun Matsumoto*, Hajime Tanuma*, Toshiyuki Azuma†, and Haruo Shiromaru*

1Department of Physics, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo 192-0397, Japan
2Department of Chemistry, Tokyo Metropolitan University, 1-1 Minami-Osawa, Hachioji, Tokyo 192-0397, Japan
3RIKEN, 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

Synopsis
We directly observed recurrent fluorescence from small carbon cluster anions C6- stored in an electrostatic ion storage ring TMU E-ring. This process, caused by the electronic transitions via low-lying electronic excited states after the inverse internal conversion (IIC), has been recently reported as fast cooling. In the present study, we succeeded in detection of emitted fluorescence. The observed lifetime was consistent with the theoretically estimated lifetime.

In an isolated environment, vibrationally excited molecules experience fragmentation, electron emission and IR radiation for the vibrational transitions. Recently, an additional rapid cooling process of emitting fluorescence has been experimentally revealed for the molecular ions stored in ion storage devices [1-3]. This emission called recurrent fluorescence is accompanied by the electronic transitions via low-lying electronic excited states after the inverse internal conversion (IIC) process, i.e., the conversion of internal energy from vibrationally excited states to electronically excited states (see Fig. 1).

Figure 1. Jablonski diagram of the molecular anions with the low-lying electronic excited state.

It should be noted that the fast cooling rates observed in the studies reported so far [1-3] was derived from the yield of the neutral products through the dissociation or electron detachment. In this study, we succeeded in first observation of the recurrent fluorescence photons emitted from the electronically excited states C2Πg+ (2.04 eV above the ground state) of C6- ions stored in an electrostatic ion storage ring, TMU E-ring.

Figure 2 shows the schematics of the experimental setup. 15 keV C6- ion bunches of typically less than 1 μs width, produced with a laser desorption ion source, were injected and stored in the TMU E-ring with a circulation period of 39.2 μs. After the storage time of typically 0.5 ms, pulsed laser light (a second harmonic light of the pulsed Nd:YAG laser, wavelength: 532 nm, energy: 9 mJ/pulse) was irradiated to excite the stored ions. We observed the delayed recurrent fluorescence from the stored C6- ions by a Pertier-cooled photomultiplier tube (PMT) for the single photon detection. In front of the PMT, four lenses, an interference filter (607 nm CWL, 80 nm FWHM), and an aperture for confining the field of view, were placed. All the optical devices and a PMT were covered with a dark box to reduce the background light.

The observed lifetime of the fluorescence was apparently longer than the intrinsic lifetime of the C2Πg+ state, 730(50) fs [4]. The rapid decrease of the fluorescence was consistent with our theoretically estimated lifetime taking account of the IC and IIC processes.

Figure 2. Schematic of the photon detection setup with the optical devices and a photomultiplier tube.

References

*E-mail: takeshi@tmu.ac.jp