Sensing ultrafast motion of a single atom encapsulated in a cage molecule by terahertz spectroscopy

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Upon the discovery of the superatom states in endohedral metallofullerenes (EMF), the superatom properties have become attractive, because ultrafast motion of the trapped atom modifies the electron density distribution in a local area of a picometer-scale. So far, the encapsulated atom position was imaged by scanning tunneling microscopy or transmission electron microscopy. However, they can image the encapsulated metal atom only in a time-averaged manner; the observation of ultrafast atom motion is very challenging because dynamical processes take place in the terahertz (THz) frequency range in a picometer-scale.

Here, we report on the THz spectroscopy of single EMF molecules by using the single molecule transistor (SMT) geometry [1], as shown in Fig. 1(a). The fullerene works as a natural cage to trap a metal atom. Using the source and drain electrodes separated by a sub-nm gap as a THz antenna, we focused the THz radiation [2] onto a single EMF molecule trapped in the nanogap electrodes. By using a blackbody THz source [3], we have observed two broad photocurrent peaks associated with the bending and stretching motions of the encapsulated single atom, as shown in Fig. 1(b). These broad peak widths are consistent with theoretical predictions [4] that the rattling motion of the Ce atom in the C_{82} cage is very anharmonic.

The present result demonstrates that the THz spectroscopy together with nanogap electrodes have attained a single-atom level spatial resolution and a picosecond time resolution.

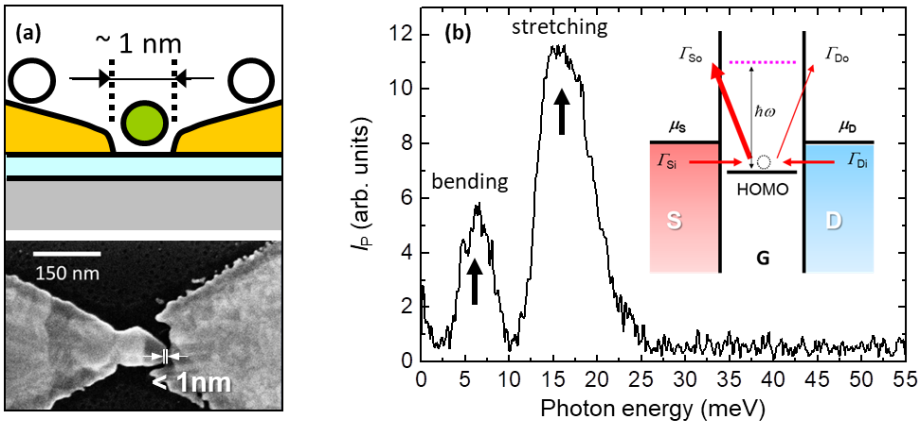

Figure 1 (a) Schematic and SEM image of the nanojunction region of a single molecule transistor (SMT). **(b)** Spectrum of the THz-induced photocurrent in a single-Ce@C_{82} SMT. The inset shows THz-induced vibron-assisted tunneling process.

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