Terahertz spectroscopy of single Ce@C_{82} molecules using sub-nm scale gap electrodes

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Terahertz (THz) spectroscopy is a powerful tool for clarifying electronic structures and vibrational dynamics of various kinds of molecules. However, it is a great challenge to greatly exceed the diffraction limit [1] and perform single molecule spectroscopy, because there is a huge size difference (a factor of ~10^5) between the THz wavelength (~100 μm) and the size of single molecules (~1 nm).

In this work, we propose a novel method for performing THz spectroscopy on single molecules. THz vibrational spectra of a single Ce@C_{82} molecule have been measured by using a single molecule transistor (SMT) geometry, which consists of a single molecule and metal nanogap electrodes, as a THz detector. The inset of Fig. 1(b) shows an SEM image of a fabricated Ce@C_{82} SMT structure. We created a sub-nm gap in a metal nanojunction by using electromigration. By using the nanogap electrode as a THz antenna, this sample structure allows us to overcome the diffraction limit and focus THz radiation onto a single molecule. Figure 1(a) shows the Coulomb stability diagram of a Ce@C_{82} SMT. The crossing pattern indicates that we capture a single molecule in the nanogap. THz signal of a single Ce@C_{82} molecule was obtained by measuring the THz-induced photocurrent in the SMT sample. Figure 1(b) shows the obtained THz spectra of a single Ce@C_{82} molecule. Broad photocurrent peaks appear around 50 cm^{-1} and 110 cm^{-1}. Although the origin of these peaks is not clear at present, we think it is related with dynamical motion of the encapsulated Ce atom in the C_{82} cage [2]. This is the first successful measurement of THz spectra of a single molecule.

Reference: [1] Y. Zhang et al, Nano Lett. **15**, 1166 (2015). [2] W. Andreoni and A. Curioni, Phys. Rev. Lett., **77**, 834 (1996).

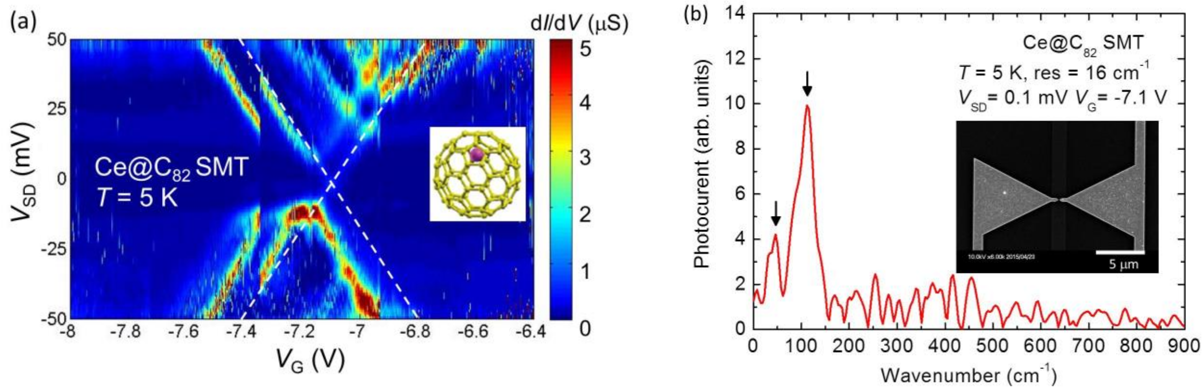

Fig. 1 (a) Coulomb stability diagram measured on a Ce@C_{82} SMT. White dash lines are eye guides for the Coulomb diamonds. (b) THz spectra measured near the charge degeneracy point (V_{SD} = 0.1 mV, V_G = -7.1 V) shown in Fig. 1(a). The resolution of the measurement was 16 cm^{-1}. The inset is an SEM image of a fabricated SMT integrated with a bow-tie antenna.