Photon-Assisted Tunneling through Single Molecules Induced by Terahertz Radiation Enhanced in the Sub-nm Gap Electrodes

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***Abstract*— We have investigated the electron transport in single C_{60} molecule transistors under the illumination of intense monochromatic terahertz (THz) radiation. By employing an antenna structure with a sub-nm wide gap, we concentrate the THz radiation beyond the diffraction limit and focus it onto a single molecule. The photon-assisted tunneling (PAT) in the single molecule transistors has been observed both in the weak-coupling and Kondo regimes. The THz power dependence of the PAT conductance indicates that, when the incident THz intensity is a few tens mW, the THz field induced at the molecule exceeds 100 kV/cm, which is enhanced by a factor of ~10^5 from the field in the free space.**

I. INTRODUCTION

Recently, electron transport through single molecules is attracting considerable attention owing to their potentiality of utilizing a variety of molecular functions for electronics [1-5]. So far, most of the works on the single molecule transport has been performed on their static properties and very little has been done on their dynamical transport. Typical energy scales in the single molecule transport lie mostly in the terahertz (THz) frequency range and interactions between THz fields and single molecules may well result in intriguing transport phenomena.

Here, we report on electron transport in single C_{60} molecule transistors (SMTs) under the illumination of monochromatic THz radiation at 2.5 THz with an intensity of a few tens mW. We have fabricated a sample structure that can focus the THz radiation onto a single molecule trapped in the nanogap electrodes. Under the THz radiation, the SMTs exhibit satellite conductance lines that arise from the photon-assisted tunneling (PAT). From the power dependence of the PAT conductance, we have found that the THz electric field induced across the nanogap electrodes exceeds 100 kV/cm, which is enhanced from its value in the free space by a factor of ~10^5.

II. RESULTS

We fabricated C_{60}-SMTs by using the electrical break junction method [6, 7]. An 8-nm-thick NiCr layer, which served as a semi-transparent backgate electrode, was deposited on a semi-insulating GaAs substrate and a 30-nm-thick Al_2O_3 gate-insulation film was grown by using the atomic layer deposition. After that, we formed thin gold nanojunctions for the source and drain electrodes on the surface of the wafer by using the shadow evaporation technique. To achieve a good coupling efficiency between the THz radiation and tunnling electrons, we employed a bow-tie antenna shape for the contacting electrodes and fixed the sample on a hemispherical silicon lens. Figure 1 shows the Coulomb stability diagrams of a C_{60}-SMT under three different incident THz intensities, P_{THz}. When the THz radiation is present, additional $\partial I_{\mathrm{SD}}/\partial V_{\mathrm{SD}}$ lines parallel to the ground state lines appear both in the single electron transport and the Coulomb blockaded regions. The energy separations between the ground state line and the satellite lines are found to be ~ ±10 meV, which agrees with the photon energy of the THz radiation (hf = 10.3 meV). When P_{THz} is increased to 55 mW, not only do the satellite peaks at ±10 meV grow in magnitude but also a new satellite peak shows up 20-meV above the ground state line, indicating that a two-photon absorption process takes place when P_{THz} is increased. In contrast, the tunnel conductance for the ground state line is reduced with increasing P_{THz}.

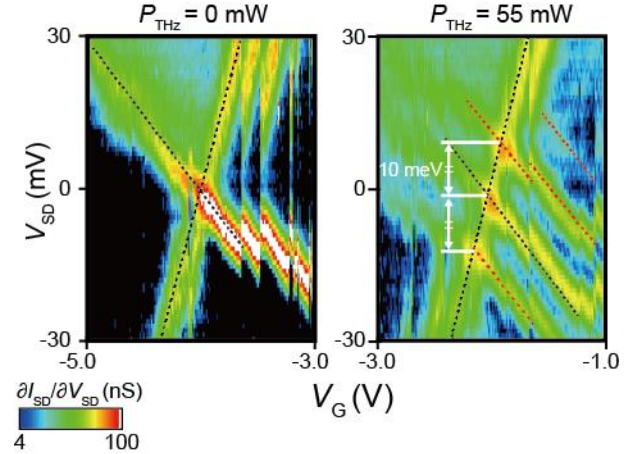

Fig. 1 Coulomb stability diagrams measured under the illumination of 2.5 THz radiation for the THz power P_{THz} = 0 and 55 mW (from left to right). Black dotted lines represent the boundaries between the transport and Coulomb blockaded regions. Red dotted lines denote the differential conductance peaks generated by the THz radiation.

III. DISCUSSION

The conductance for the N-photon PAT process, G_N, is known to be proportional to $J_N^2(\alpha)$, where J_N is the N-th order Bessel function and $\alpha \equiv eV_{\mathrm{THz}}/hf$ [8]. V_{THz} is the THz voltage induced across the nanogap. The ratio between the ground state conductance, G_0, and the conductance of the N-th satellite, G_N, is, then, given by $G_N/G_0 = J_N^2(\alpha)/J_0^2(\alpha)$. Figure 2 plots the conductance ratios G_1/G_0 and G_2/G_0 as a function of P_{THz}. As seen in Fig. 2, the experimental data for G_1G_0 are indeed well fitted by $J_1^2(\alpha)/J_0^2(\alpha)$, which also supports that the satellite channels are created by the PAT effect. Note that α is in the order of unity when $P_{\mathrm{THz}} \geq 36$ mW. Since the nanogap distance, d, is comparable to the size of the C_{60} molecule (diameter ~ 0.7 nm), $\alpha \equiv eV_{\mathrm{THz}}/hf \sim 1$ indicates a remarkable fact that the THz field in the gap $E_{\mathrm{THz}} = V_{\mathrm{THz}}/d \sim hf/ed > 100$ kV/cm. Since E_{THz} in the free space is in the order of a few V/cm in the experimental condition used in this work, this means that the THz field at the molecule is enhanced by a factor of ~10^5 from its free-space value by the plasmonic effect of the metal nanogap electrodes. This large field enhancement is in the order of λ/d (λ: THz wavelength, d: nm-scale gap width).

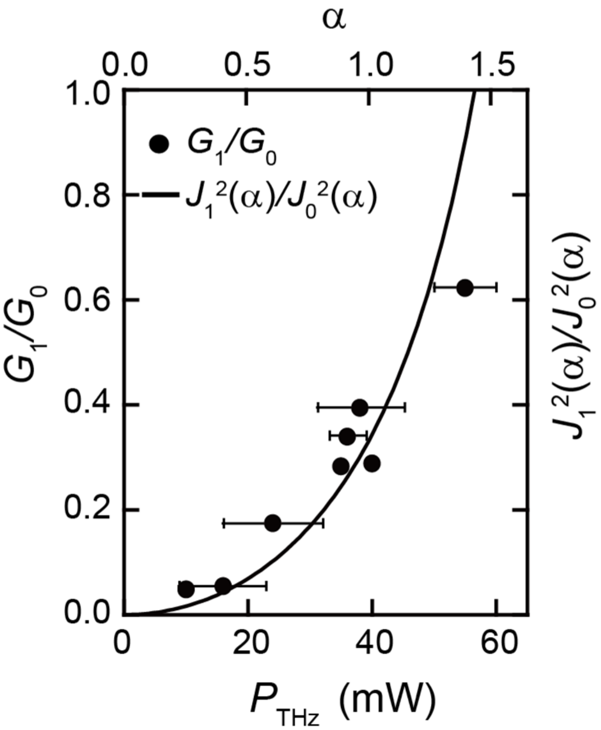

Fig. 2 The P_{THz}-dependence of G_1/G_0 (black dots). The black curve indicate $J_1^2(\alpha)/J_0^2(\alpha)$ as a function of α

IV. SUMMARY

In summary, we have investigated quantum charge transport through single C_{60} molecules under the illumination of intense monochromatic THz irradiation. We have achieved highly efficient focusing of the THz radiation onto single molecules and observed the PAT in the SMTs. Intense THz fields above 100 kV/cm are induced in the sub-nm gap of the electrodes by the plasmonic effect. The THz field enhancement factor of the system reaches as high as ~10^5.

ACKNOWLEDGEMENT

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