Colossal terahertz nonlinearity of angstrom-sized infinite gaps

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***Abstract*—We observed funneling of terahertz waves through angstrom gaps formed by metal-graphene-metal hybrid structures in low-field regime, which is almost completely blocked in high-field regime. The unprecedented transmission nonlinearity of 97% was induced by electron tunneling across the angstrom gap.**

I. INTRODUCTION

METAL-graphene-metal hybrid structures provide the smallest possible optical gaps, through which quantum tunneling routinely occurs, making this system an ideal platform for quantum plasmonics. Recently, there has been considerable interest in the development of sub-nanometer gap fabrication techniques using a single layer graphene as spacer between two metal films or particles [1, 2]. The simple vertical stacking of metal-graphene-metal structures, however, is difficult to be applied for millimeter waves because of typical metal film thickness and particle size (~100 nm) limited by evaporation and synthesis techniques. To study the angstrom gap using electromagnetic waves in broad wavelength ranges, one is required to manufacture angstrom gaps of an infinite length in the sense of being much longer than any of the wavelengths of the incident light. Ultra-long angstrom gaps enable electromagnetic waves to squeezing through the gap without any strays and allow background-free probing. In this work, we manufactured five millimeter long angstrom gaps formed by metal-graphene-metal structures, through which terahertz waves funnel. Our sample enables quantum plasmonics of long wavelength light with giant nonlinearities.

II. RESULTS

The angstrom-sized gaps of metal-graphene-metal were made by chemical vapor deposition system and simple adhesive tape based planarization [3] as shown in Figure 1. First, we prepared a 300-nm-thick copper film patterned with an array of rectangular apertures of 5 mm by 0.2 mm onto a quartz substrate using photolithography (Fig. 1(a)). A single layer graphene was grown on the patterned copper film by chemical vapor deposition (Fig. 1(b)). Next, a secondary copper layer was deposited on the same sample by thermal evaporation (Fig. 1(c)) and adhesive tape was applied to the surface to peel off the second copper layer selectively (Fig. 1(d)). The final structure can be considered as an infinitely long slit array with a period of 0.2 mm for the terahertz spot size of about 2.5 mm. The effective gap size of the structure is about 3 Å [1, 2].

We performed terahertz time-domain spectroscopy using both oscillator- and amplifier-based terahertz system. A femtosecond Ti:sapphire laser operating at 80 MHz repetition rate is used to illuminate a biased GaAs emitter for generation of low power terahertz pulses with a maximum electric field of 30 V/cm [4]. For high power terahertz generation, a 1 kHz Ti:sapphire regenerative amplifier system is used to illuminate a prism-cut $LiNbO_3$ crystal with pulse-front-tilted efficient optical rectification [5]. From the setup, the maximum electric field is about 200 kV/cm [6]. The strength of the electric field is controlled by a pair of wire grid polarizers allowing us to change the field from 10 to 200 kV/cm. In both setups, the transmitted terahertz electric fields are collected with parabolic mirrors and detected by electro-optic sampling technique using <110> oriented GaP and ZnTe crystals [7, 8].

Figure 2(a) shows temporal profiles of terahertz pulses transmitted through the sample for different incident electric fields, divided by the maximum field amplitudes transmitted through quartz substrate. For low-intensity terahertz pulses ($E_{0,\max} \sim 30$ V/cm), the normalized transmitted amplitude is about 1% at the peak, leading to a field enhancement of 7000 extracted by Kirchhoff integral formalism [9]. We note that the direct transmission through the copper film was subtracted. As we increase the incident terahertz field up to 200 kV/cm, the normalized amplitude decreases down to 0.16%. Figure 2(b) presents the normalized transmittance at the peak in time domain, as a function of the incident terahertz electric field. Ninety-seven percent of nonlinearity was clearly seen in the normalized transmitted power.

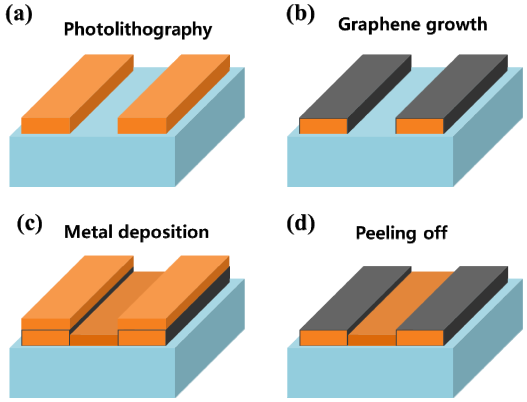

Fig. 1. Schematic of fabrication process for millimeters long, angstrom sized gaps formed by copper-single layer graphene-copper composite. (a) A patterned copper layer with rectangular structures with 5 mm by 0.2 mm is prepared by photolithography. (b) A single layer graphene is directly grown on the patterned copper film by chemical vapor deposition. (c) The second copper layer is deposited by thermal evaporation. (d) An adhesive tape is applied to the sample to peel off the second copper layer selectively.

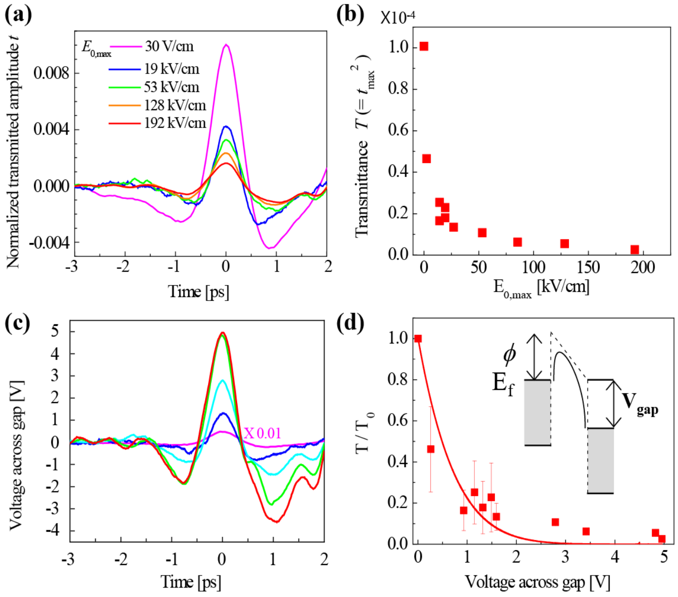

Fig. 2. (a) Time traces of transmitted terahertz electric field through angstrom gaps of copper-graphene-copper, normalized by the substrate transmission for different terahertz electric fields. (b) Maximum normalized transmittance as a function of the incident terahertz electric field. (c) Transient voltages across the angstrom gaps for different incident terahertz electric fields. (d) Experimental (square dot) and calculated (solid line) nonlinear response T/T_0 of the angstrom gap sample as a function of the induced voltage for the gap size w of 3 Å and barrier height ϕ of 3 eV

To understand the origin of the nonlinear response, we plotted the induced voltage across the gap by multiplying the incident electric field with field enhancement factor. The maximum voltage across the gap is changed from 5 mV to 5 V when the incident electric field increases from 30 V/cm to 192 kV/cm (see Fig. 2(c)). The induced voltage is sufficiently strong to deform the quantum mechanical potential barrier of the angstrom gap and facilitate electron tunneling across the gap. We calculated the tunneling current density J and conductivity σ across the angstrom gap under the applied voltage V by assuming a square quantum barrier modified by an image potential as shown in the inset of Fig. 2(d) [10]. The applied voltage corresponds to the funneling-induced voltage across the angstrom gap. Based on a quantum corrected plasmonic model of dielectric constant [11] and modal expansion [12], the change of transmittance T/T_0 was calculated as a function of the applied bias voltage for the effective gap size of 3 Å, the barrier height of 3 eV and high frequency dielectric constant of three [13] (see Fig. 2(d)). The calculated result is well matched with the experimental data (red dots). As a result, it was well explained that the giant nonlinearity originates from terahertz funneling-induced electron tunneling across the angstrom gap, which modifies the effective dielectric constant of the fictitious material inside the gap [11].

III. SUMMARY

We observed 97% decrease of normalized transmittance in a few millimeter long, angstrom-sized gaps at terahertz frequencies. The colossal nonlinearity originates from an intense terahertz funneling facilitating electron tunneling across the gap. Our work opens up long wavelength quantum plasmonics and angstrom optics.

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