Supporting Information

Metamaterial-free Flexible Graphene-enabled Terahertz Sensor for Pesticide Detection at Bio-interface

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Fabrication of graphene-based THz sensor on silicon substrate

Ti with a thickness of 10 nm and gold with a thickness of 200 nm was deposited on the silicon substrate in sequence through vacuum evaporate plating technology. Then we carefully attached polyimide tape onto the surface of gold without bubbles. The thicknesses of polyimide tape are 50 μm, 60 μm, and 80 μm, respectively. Graphene monolayer was then transferred onto the surface of polyimide, forming a graphene-based THz sensor.

Fabrication of flexible graphene-based THz sensor using conductive tape

Graphene monolayer was transferred onto the surface of a thin polyimide film (50 μm), and a conductive tape was then carefully attached onto the other surface of this film.

Fabrication of flexible graphene-based THz sensor using gold deposition

Ti with a thickness of 10 nm and gold with a thickness of 200 nm was deposited on the polyimide film in sequence through vacuum evaporate plating technology. Then we carefully transferred graphene monolayer onto the surface of this film, forming a flexible graphene-based THz sensor.

Simulation process

In this work, we firstly selected perfect electrical conductor (PEC) as back-reflector as most metal performs like PEC in THz band. Then we defined a dielectric material (aiming to represent polyimide film), where the refractive index was set as 1.7 and the extinction coefficient was set as 0.05. The parameters of graphene are shown in Table S1.

Simulated sensing performance of graphene sensors with different thicknesses of polyimide

Simulated sensing performance of graphene sensors with different thicknesses of polyimide is demonstrated in **Figure S1**. As shown in **Figure S1a**, the absorbance of graphene-based THz sensor decreases from 97% to 83% with the Fermi level reduces from 100 meV to 10 meV (polyimide thickness was set as 60 μm). Also the absorbance of graphene THz sensor with an 80-μm-thick polyimide decreases from 100% to 92% with the Fermi level reduces from 100 meV to 10 meV (**Figure S1b**). Compared with the simulated results in **Figure 3**, we find that with the decrease of Fermi level, the graphene sensor with polyimide thickness of 50 μm shows the largest drop of absorption value. Therefore, the graphene sensor with a polyimide thickness of 50 μm presents the best sensitivity among the three.

Figures

Figure S1. Simulated absorbance spectra of graphene sensors with the Fermi level ranging from 10 meV to 100 meV. (a) 60-μm-thick polyimide, (b) 80-μm-thick polyimide.

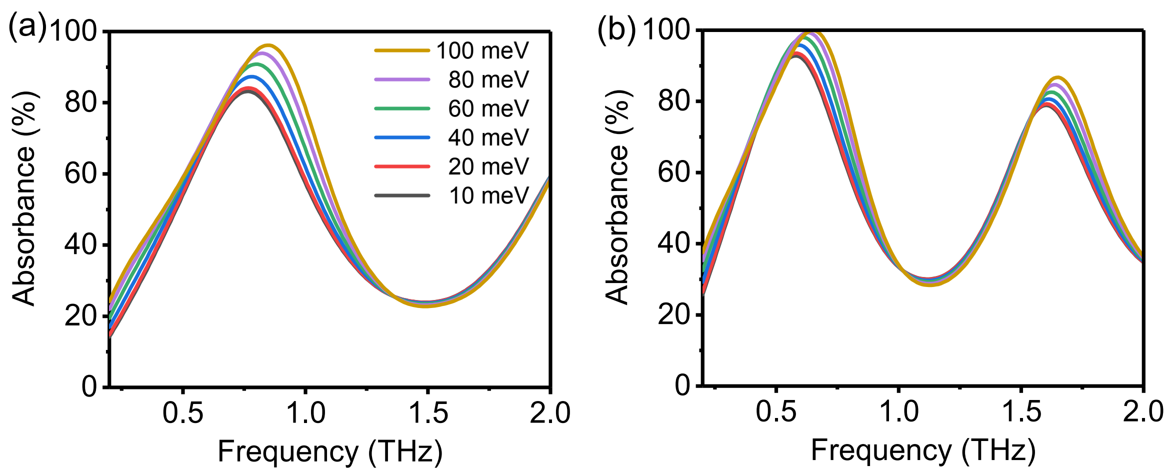

Figure S2. Simulated spectrum of graphene-based sensor operating at mid-infrared regime with a polyimide thickness of 5 μm.

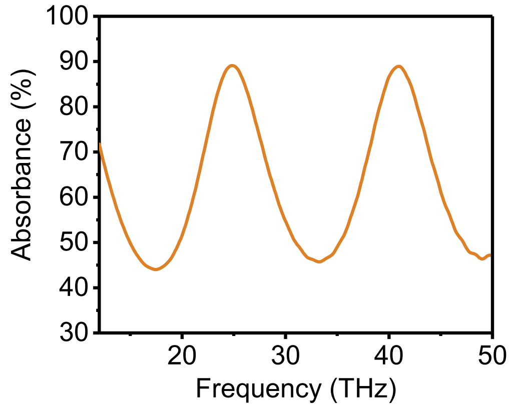

Figure S3. Measured THz spectra of graphene-based THz sensor with different layers of graphene.

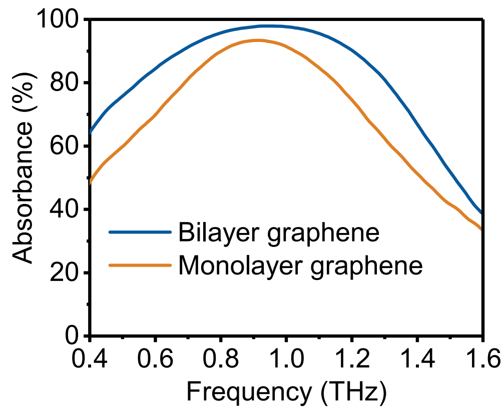

Figure S4. (a) Photograph of conductive tape. (b) SEM image of conductive tape.

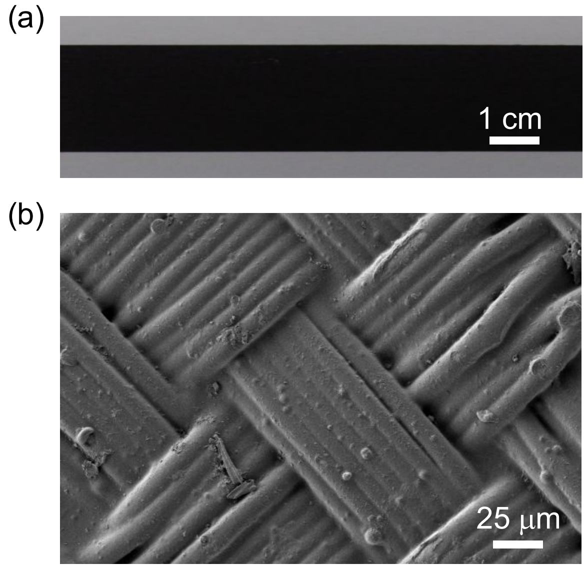

Figure S5. Measured THz spectra of polyimide film with different thicknesses on gold mirror.

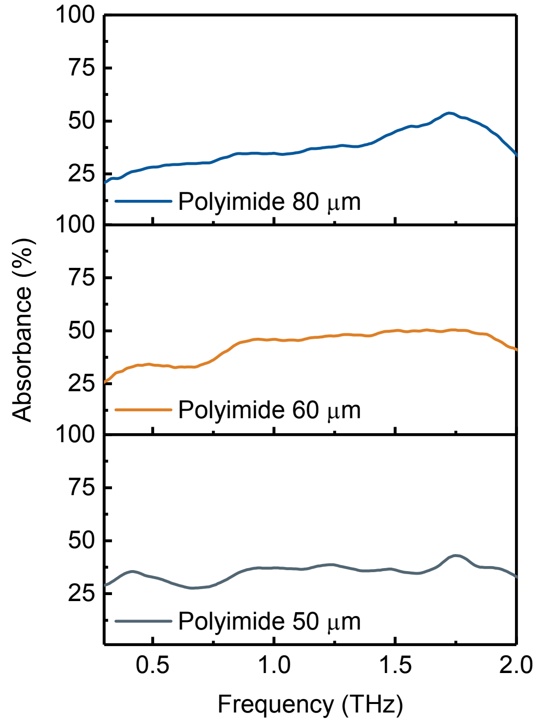

Figure S6. Measured THz time domain curves of graphene sensors with varying thicknesses of polyimide.

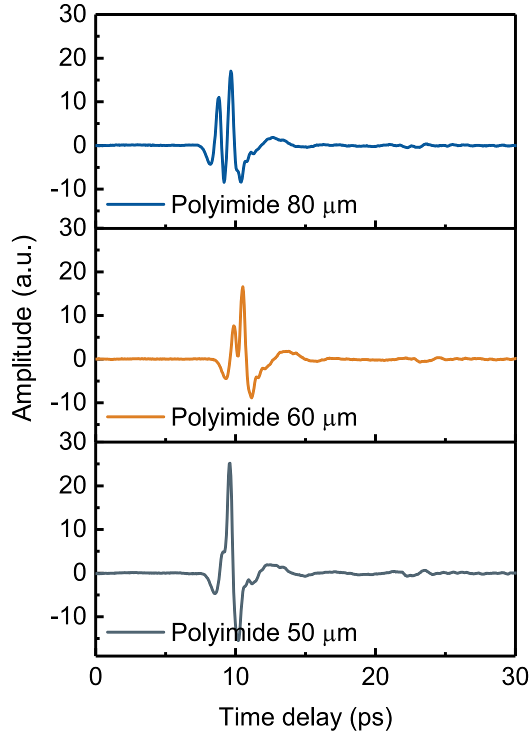

Figure S7. Absorption curve of lactose pellet (1.2 mm, 50% polyethylene and 50% lactose) ranging from 0.2 to 2.0 THz.

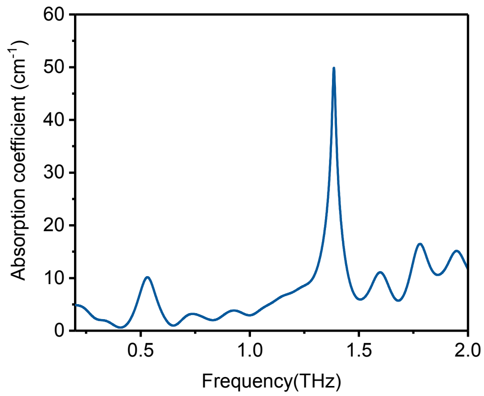

Figure S8. Simulated absorbance spectra of graphene sensors with different layers of graphene. The thickness of polyimide in this simulation is 50 μm.

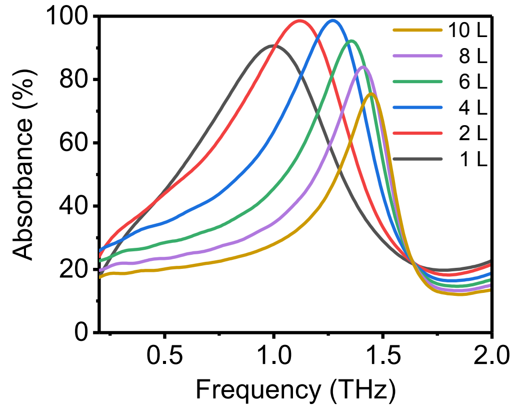

Figure S9. Simulated absorbance spectra of graphene sensors ranging from 0.2 to 10.0 THz. The thickness of polyimide in this simulation is 50 μm.

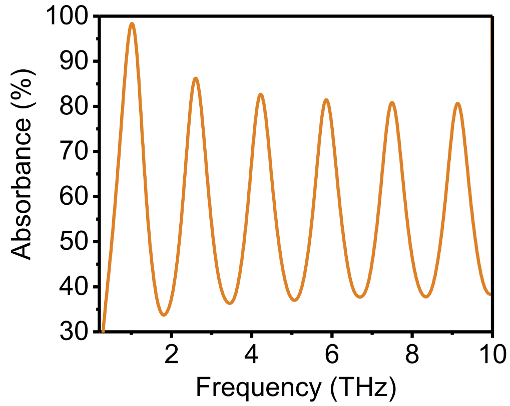

Figure S10. THz image of a polyimide-gold integrated device.

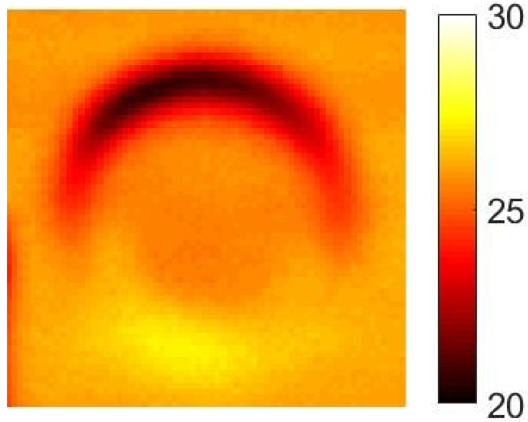

Figure S11. Reversibility of the THz sensor.

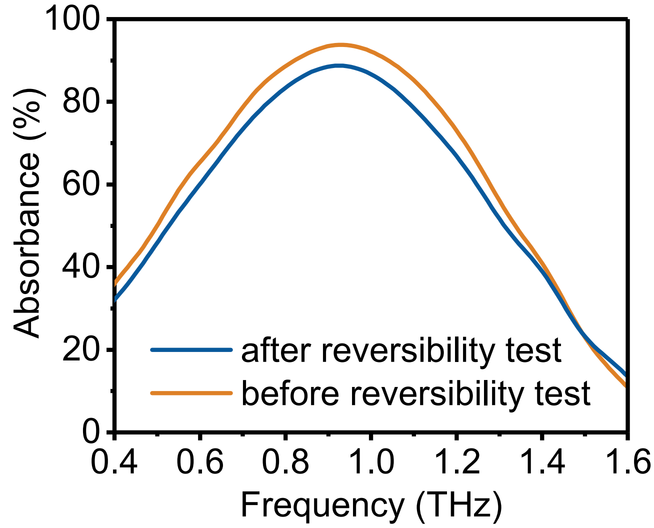

Figure S12. Experimental absorption spectrum for graphene sensor (conductive tape as back-reflector) with/without chlorothalonil. The concentration of chlorothalonil was 0.60 mg/L.

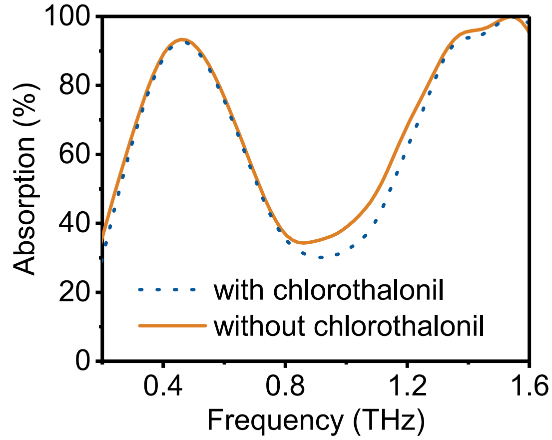

Table S1. Parameters of graphene in the FDTD simulation process.

Tolerance	0.005
Max coefficients	20
Scattering rate (eV)	0.00099
Chemical potential (meV)	10-100 (Fermi level)
Temperate (K)	300
Conductivity scaling	1-2 (Layers of graphene)