Electric Supplementary Information

Suspected graphene-based optical interferometric surface stress sensor for selective biomolecular detection

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Figure S1. Schematic diagram describing the low-pressure dry-transfer process for depositing CVD-grown graphene onto a Si substrate with pre-patterned nanocavities. Single-layer graphene was deposited by CVD onto Cu foil. (a) A polymethyl methacrylate (PMMA) solution was spin-coated onto the graphene/Cu foil. (b) The Cu layer was removed by etching with an FeCl$_3$ solution while the PMMA/graphene sheet was supported by a polydimethylsiloxane (PDMS) block, followed by rinsing in deionized water. (c) The PDMS/PMMA/graphene was transferred onto a thermal-SiO$_2$/Si substrate with shallow cavities created by reactive ion etching in a vacuum chamber. The graphene-contacted substrate was heated on a hotplate to increase the temperature above the glass-transition temperature of PMMA, leading to strong adhesion. (d) The supporting PMMA layer was removed using an N-Methyl-2-pyrrolidone-based solvent at 60 °C. (e) Finally, the suspended graphene was dried by CO$_2$ supercritical drying.
Figure S2. Optical interference measurements for the graphene drums with diameters of \( \leq 10 \, \mu m \) were performed using microspectroscopy. White light from a xenon lamp (Asahi Spectra, LAX-C100) was used to illuminate the sensor chip via a reflection microscope (Olympus, BX51). A visible spectrometer (Ocean Optics, USB2000) was attached to the microscope using an optical fiber with a core diameter of 100 \( \mu m \). The magnification of the objective lens was 50\( \times \), and the spot diameter was 2 \( \mu m \), which was a sufficiently small measurement area for the fabricated graphene drum.
Figure S3. Cross-reactivity between anti-bovine serum albumin (BSA) and human serum albumin (HSA) was evaluated by fluorescence observations. (a) Two types of protocols for treating BSA antigen with anti-BSA antibody (positive control) and treating HSA with anti-BSA antibody are shown. We prepared two substrates modified with anti-BSA antibody (100 μg/mL) and casein for blocking (5% skimmed milk). The anti-BSA-modified chips were immersed in a 100 μg/mL BSA antigen solution and a 100 μg/mL HSA solution, followed by treatment in a 100 μg/mL fluorescein isothiocyanate (FITC)-conjugated anti-BSA antibody solution as a secondary antibody. After each step, the chip was washed with phosphate-buffered saline (PBS). The treated chips were immersed in PBS, and (b) bright field observation and (c) fluorescence observation were performed. Fluorescence was observed only in the BSA antigen-treated chip, indicating that there is no cross-activity between anti-BSA and HSA.
Figure S4. Typical reflection spectra at the saturation of a 6-µm-diameter drum corresponding to BSA concentration; (a) 1 µg/mL, (b) 100 ng/mL, (c) 10 ng/mL, and (d) 1 ng/mL. The fitting curve was drawn using Equation 1 with the air gap as a parameter. The gap change corresponds to the graphene deflection.