Atomic Emission Spectroscopy of Electrically-Triggered Exploding Nanoparticle Analytes on Graphene/SiO₂/Si Substrate

Siyang Liu, Myungji Kim, and Hong Koo Kim

Department of Electrical and Computer Engineering and Petersen Institute of NanoScience and Engineering, University of Pittsburgh, Pittsburgh, PA 15261 Siyang.Liu@pitt.edu

We present a device technology that promises chip-scale atomic-emission spectroscopy for elemental analysis of analyte materials. In the proposed structure, analytes are placed on top of a graphene/SiO₂/Si substrate and are atomized for atomic luminescence under electrical excitation. Here the graphene layer is designed to serve as an electron-transparent conducting electrode of a graphene/oxide/semiconductor (GOS) capacitor structure (Figure 1(a)).

Under proper voltage pulses applied to the gate (graphene anode), the thin insulating layer (SiO₂) breaks down forming nanoscale leakage channels. (Besides the breakdown-induced leakage channels we also fabricated nanohole arrays in the SiO₂ layer by employing nanolithography (electron-beam or focused-ion beam lithography: Figure 1(c)). Inversion (or accumulation) electrons induced at the SiO₂/Si interface then emit into the void channels and ballistically travel into the anode side, where analytes sit on graphene electrode. Being transparent to impinging electrons the graphene electrode allows bombardment of nanoparticle analytes (CdSe quantum dots in the present work) with kinetic electrons. The nanoparticle analytes accumulate charges while being heated. When the Coulomb repulsion within charged particles becomes stronger than the binding force, the nanoparticles explode, atomizing the compound semiconductor analytes. This explosive fragmentation is expected to produce atoms in various excited states. The excited atoms then relax producing characteristic luminescence. The luminescence spectra are captured and analyzed with a CCD-based optical spectrum analyzer. Figure 1(b) shows an atomic spectrum measured from CdSe quantum dots (monolayer) placed on top of a graphene/SiO₂ (~10nm thick)/p-Si (GOS) capacitor structure under pulsed bias. For voltage pulses (~50 V amplitude and 1 µs - 1 ms pulse width) applied to the gate of the GOS structure, atomic luminescence from Cd was observed at 509 nm, 480 nm, and 468 nm. Atomic luminescence from silicon (590 nm and 517 nm) and oxygen (644 nm) was also observed.

¹ S. Srisonphan, Y. S. Jung, and H. K. Kim, Nature Nanotechnology 7, 504–508 (2012).

² M. Kim and H. K. Kim, J. Appl. Phys. 118, 104504 (2015).



Figure 1: Atomic emission spectroscopy enabled by electrically-triggered explosion of nanoparticle analytes on graphene/SiO₂/Si substrate. (a) Schematic of device structure and system set-up for emission detection. (b) A measured spectrum of atomic emission from CdSe quantum dots and SiO₂/Si substrate under 50 V-10 μ s positive pulses. (c) SEM image of a nano-hole array formed in SiO₂/Si by electron-beam lithography and plasma etching. Scale bar, 500 nm. (d) Optical micrograph of sample surface before explosion. Red lines show the boundary of graphene layer. (e) Optical micrograph of atomic emission during explosion. Scale bars in (d) and (e) are 200 μ m.