Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2015

Supporting Information

Raman Enhancement Effect on Thin GaSe Flake and Its Thickness

Dependence

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Figure S1. a. STEM image of a thin GaSe. Insets are the magnified image (top right) and intensity profile along the solid line (bottom right). b. Schematic illustrations of the structures of β -polytype (left) and ϵ -polytype GaSe (right), respectively. (adopted from reference 1).



Figure S2. EDX spectrum of the GaSe. The atomic ratio of Ga:Se is calculated about 1:1.



Figure S3. AFM images of different GaSe layers. The height of the flakes are (a) 3.5 nm, corresponding to ~4 layer GaSe, (b) 5.0 nm, corresponding to ~6 layer GaSe, (c) 13.5 nm, corresponding to ~16 layer GaSe, respectively.



Figure S4. High-resolution STEM image of ultrathin GaSe (less than 4 layers). Inset is the SAED.



Figure S5. Low frequency region of Raman spectra of CuPc deposited on the SiO_2/Si and the GaSe flakes with different thickness.



Figure S6. Solid line is the calculated relative electromagnetic enhancement from interference effect. Data points are the experimental relative enhancement factors (from Figure 4b of manuscript) as a function of layer number of GaSe flakes for several typical vibration modes of CuPc. The results demonstrate that the interference has little influence on the Raman enhancement.



Figure S7. Raman spectra of 1nm CuPc deposited on the thin GaSe flake (red line), thick GaSe flake (blue line) and the SiO_2/Si substrate (black line) with 633 nm excitation.



Figure S8. PL spectra for pristine GaSe samples (monolayer, few-layer, multilayer) and corresponding GaSe samples with 1 nm CuPc coating, under the 514 nm laser excitation at room temperature.

Reference

X. Li, L. Basile, M. Yoon, C. Ma, A. A. Puretzky, J. Lee, J. C. Idrobo, M. Chi, C.M. Rouleau, D. B. Geohegan, and K. Xiao., *Angew. Chem. Int. Ed.*, 2015, 54, 2712 –2717.