SUPPORTING INFORMATION

Raman analysis of bilayer graphene film prepared on commercial Cu(0.5 at% Ni) foil

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Ni surface concentration calculation using surface segregation models

Part of the modified Darken equations, which defines the rate of surface concentration build-up of dope element 1 (e.g. Ni in dilute Cu(Ni) foil), is[1,2]

$$\frac{\partial X_1^S}{\partial t} = \frac{M_1 X_1^{B_1}}{d^2}\left[\Delta G + RT \ln \frac{X_1^{B_1}\left(1-\frac{X_1^S}{X^M}\right)}{\frac{X_1^S}{X^M}(1-X_1^{B_1})}\right] \tag{1}$$

where X_1^S is the surface concentration of the segregating dope element 1, X_1^B is the bulk concentration, X^M is the maximum surface concentration < 100 at%, M_1 is the mobility, $M = D/RT$ where D is the diffusion coefficient in Arrhenius equation, d is the thickness of the segregated layer, R is the gas constant and T is the temperature.

In dilute system the segregation energy (ΔG) can be approximated to segregation enthalpy (ΔH) by [3,4]

$$\Delta G \approx \Delta H = \left(\frac{\Delta Z}{Z}\right)\left(\Delta H_B^{sub} - \Delta H_A^{sub}\right) \tag{2}$$

where Z is bulk coordination number (Z = 12 for Cu crystal), ΔZ is a difference in coordination number between bulk and surface (ΔZ = 4, 3 and 2 for Cu(001), Cu(111) and Cu(101) respectively), ΔH^{sub} is heat of sublimation for element A and B ($\Delta H_{\mathrm{Cu}}^{sub} = 339.3$ kJ/mol and $\Delta H_{\mathrm{Ni}}^{sub} = 430.1$ kJ/mol)[5].

In Eq. S1, the rate of surface concentration build-up of dope element 1, at equilibrium, $\partial X_1^S / \partial t = 0$, and Eq. S1 reduces to the Langmuir–McLean equation[1,6]:

$$\frac{X_1^S / X^M}{1 - X_1^S / X^M} = \frac{X_1^B}{1 - X_1^B} \exp\left(\frac{-\Delta G}{RT}\right) \tag{3}$$

In Eq. S3, substituting parameters:

i. ΔG (obtained from Eq. S2 for Cu(001), Cu(111) and Cu(101)),
ii. $X_1^{B_1} = 0.5$ at % and
iii. X^M = 0.25, 0.33 and 0.50 for Cu(001), Cu(111) and Cu(101) respectively[1,6],

the temperature dependence of the Ni surface fractional concentration in dilute Cu(0.5 at% Ni) foil was obtained as shown in Fig. S1.

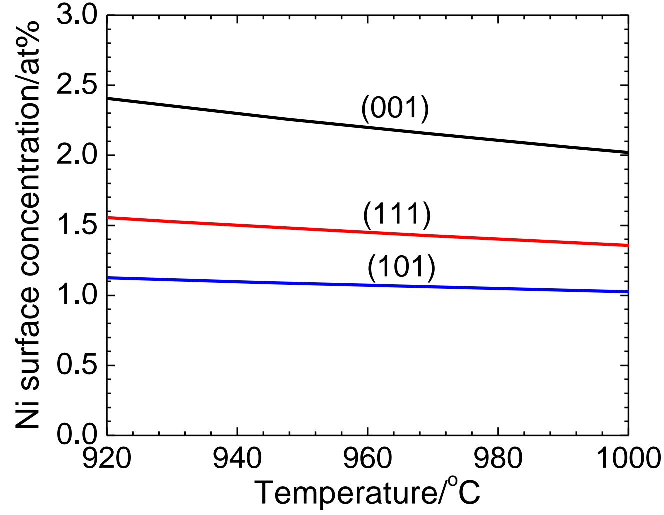

Figure. S1. The temperature dependence of the Ni surface concentration in dilute Cu(Ni) alloy obtained with Langmuir–McLean equation.

In brief, at isothermal CVD growth temperature of 980 °C the Ni surface concentration in dilute Cu(0.5 at% Ni) foil is $X_{\mathrm{Ni}}^{S} = 2.1$, 1.4 and 1.1 at% for Cu(001), Cu(111) and Cu(101) surfaces respectively. Therefore, at 980 °C, a surface layer of a dilute Cu(0.5 at% Ni) foil will have about 98.5 at% Cu and 1.5 at% Ni.

Raman data

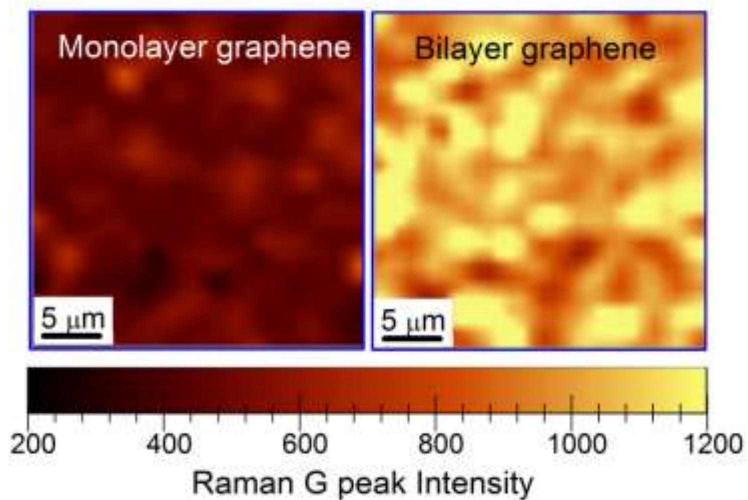

Figure. S2. The G peaks intensities mapping over 30 μm^2 area for monolayer and bilayer graphene films transferred on SiO_2/Si substrate.

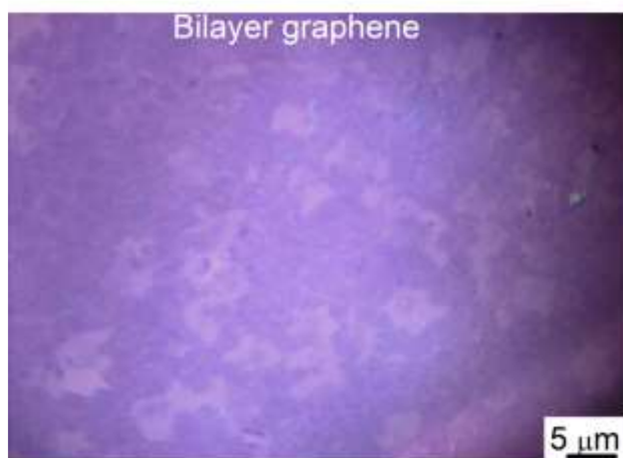

Figure. S3. The Raman optical microscope image of bilayer graphene film transferred onto 300 nm thick SiO_2/Si substrate.

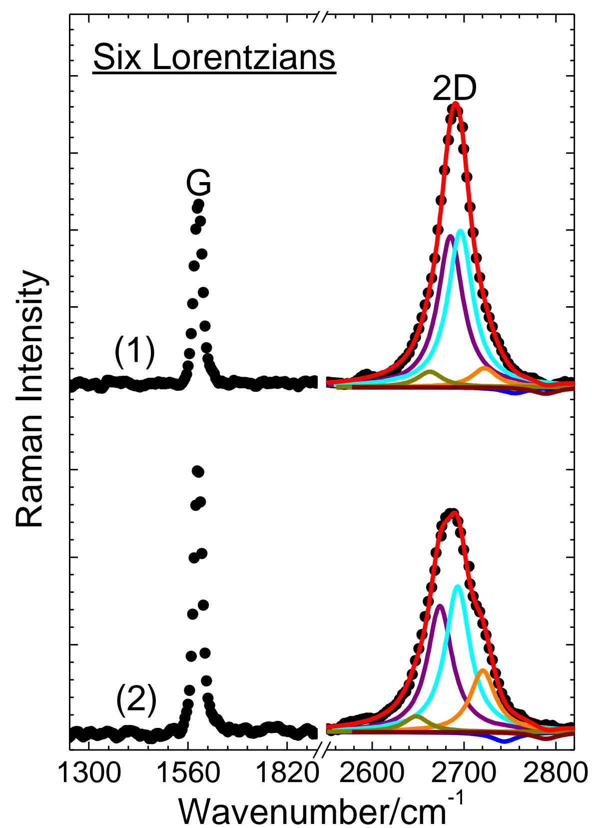

Figure. S4. The Raman spectra from two different spots of bilayer graphene film transferred onto SiO_2/Si substrate. The solid lines are Lorentzian fits of 2D peaks.

TOF-SIMS data

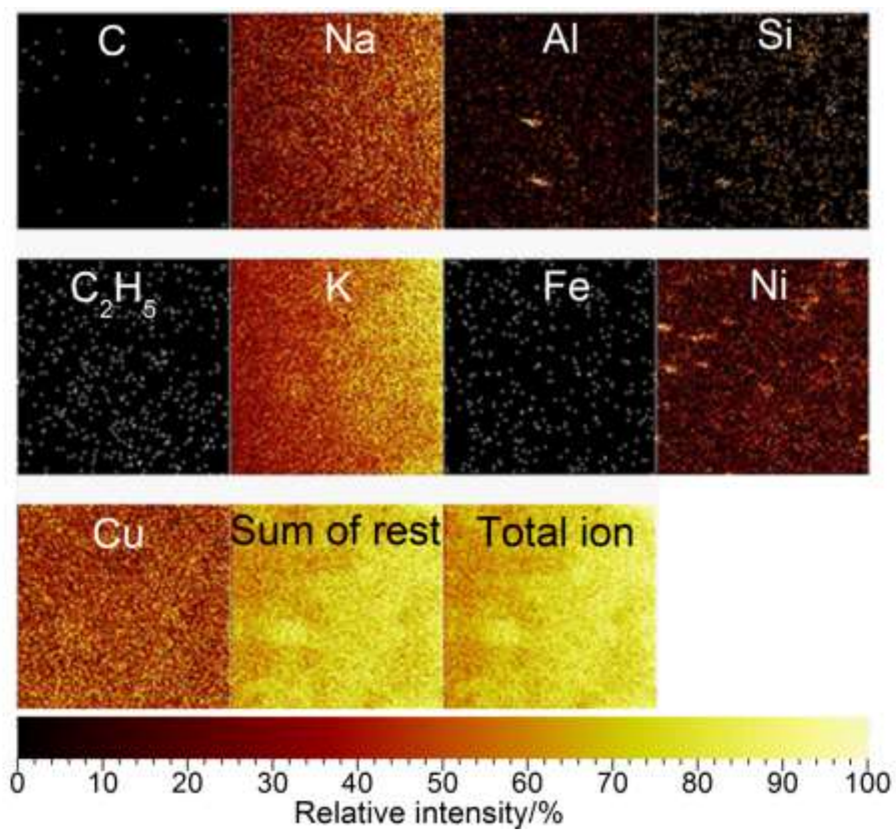

Figure. S5. The map images of TOF-SIMS secondary ion intensities measured from a dilute Cu(0.5 at% Ni) surface after surface cleaning for 180 s with ion sputtering.

S1. References

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