Epitaxial graphene on silicon substrates

Graphene, a 2D honeycomb network of sp^2-bonded carbon atoms, has been attracting considerable attention as a material for electronics as well as for photonics. Graphene expresses multifunctionality in response to the breakage of the time-reversal symmetry of its honeycomb structure [1], as conceptually described in Fig. 1. Moreover, conventional lithographic techniques for Si-ULSI are easily applicable to graphene owing to its planar structure, in contrast to the structure of carbon nanotubes. Graphene is therefore one of the most promising materials to realize the next-generation multifunctional integrated circuits can be the successor to Si.
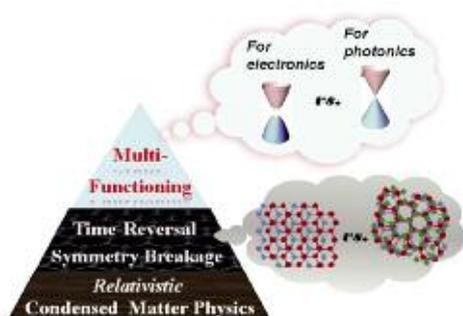

Fig. 1. Schematic of expression of multifunctionality of graphene.

Methods of growing large-scale epitaxial graphene on suitable substrates should, however, be developed in order for graphene to be the successor to Si. Among various epitaxial methods, the epitaxy of graphene on SiC bulk crystals has been considered the most promising technology. The epitaxial graphene method on SiC bulk crystals can produce high-quality films, which can be directly used for devices. In contrast, chemical-vapor-deposition-grown graphene on metal substrates needs to be transferred to insulating substrates for device applications. Unfortunately, however, epitaxially grown graphene on SiC bulk crystals has a crucial drawback: its high production cost.

To solve this drawback, we have chosen Si as the suitable substrate for the epitaxy of graphene. Namely, we have attempted and in fact succeeded for the first time in the epitaxy of graphene on Si (GOS) [2,3], as shown in Fig. 2. In fabricating GOS, a SiC thin film is grown by gas-source molecular beam epitaxy using monomethylsilane, followed by high-temperature annealing to sublimate silicon atoms

from the SiC surface for graphitization [2,3]. The GOS fabrication processes were confirmed by C 1s core level photoelectron spectroscopy to probe the formation of sp^2-bonded carbon atoms and by Raman-scattering spectroscopy to probe the network consisting of sp^2-bonded carbon atoms. Finally, cross-sectional transmission electron microscopy (XTEM) clearly images the planar structure of GOS. It can thus be concluded that graphene can definitely be grown on a Si substrate.

Field-effect transistors using GOS as a channel can be readily fabricated by standard lithography of Si, and the highest carrier mobility has been estimated to be 6,000 cm^2/Vs [4]. This high carrier mobility clearly demonstrates the availability of GOS for the next-generation devices. Furthermore, the photoresponsivity of GOS using the field effect has recently been demonstrated. GOS is thus a promising electronic and photonic material, as anticipated.

Very intriguingly, the interface properties and therefore the electronic properties of GOS can be tuned in terms of the crystallographic orientation of the Si substrate (Fig. 3) [5]. The tunability of the interface between graphene and the SiC thin film is clearly demonstrated by high-resolution C1s core level photoelectron spectroscopy co-performed at **BL23SU**, as shown in Fig. 3(a). The main features of graphene and SiC are observed in the spectra of graphene on both Si(111) and Si(100). In contrast, a peak due to the buffer layer, which is partially bonded to the underlying SiC top surface, is only observed in the spectrum of graphene on SiC(111)/Si(111).
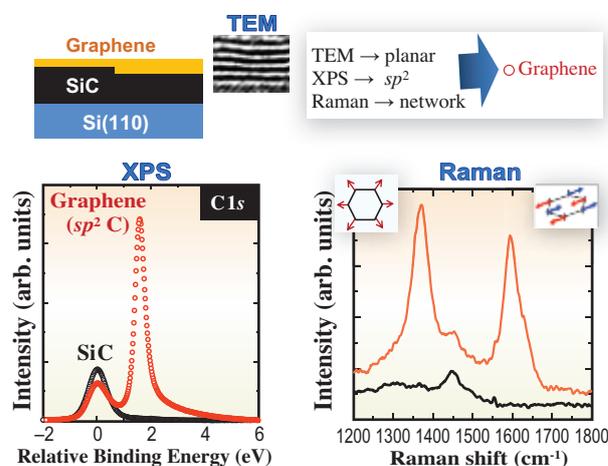

Fig. 2. C1s core level photoelectron spectra of graphene, XTEM image and Raman spectra probing epitaxial processes in GOS.
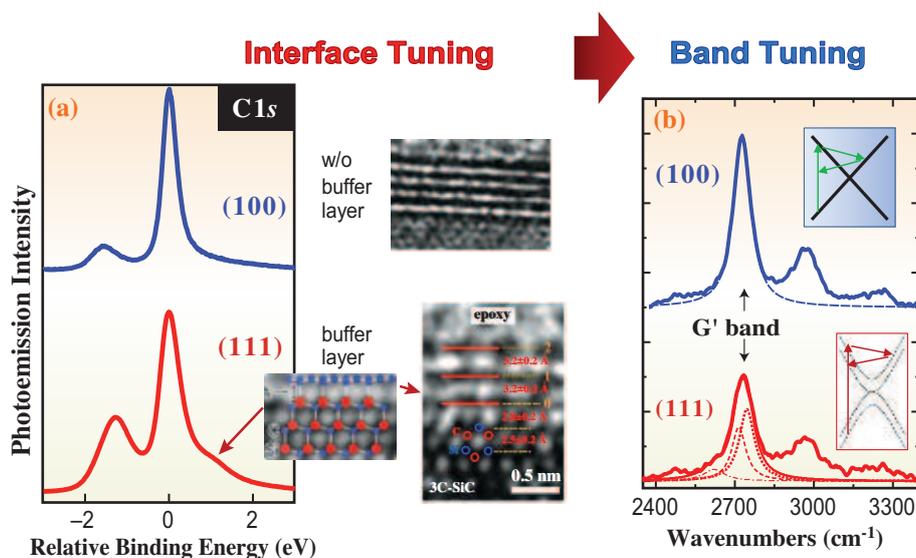
Interface Tuning → **Band Tuning**

Fig. 3. **(a)** C1*s* core level photoelectron spectra of graphene and XTEM images, and **(b)** Raman spectra, revealing the mechanism of the expression of multifunctionality.

This result of the C1*s* core level spectroscopy is validated by XTEM (Fig. 3(b)). In GOS using Si(111) as the substrate, the buffer layer is clearly imaged by that of XTEM, while the absence of the buffer layer is confirmed by XTEM in GOS using Si(100) as the substrate. The tunability of the interface structure of GOS affects the electronic structure of GOS. In Fig. 3(b), the G' bands in the Raman spectra are shown for graphene on Si(111) and Si(100). On Si(111), the G' band is multiply split as a result of the coupling between photons and multiple electronic bands. In contrast, the G' band possesses a single component in the spectra of graphene on Si(100), due to the coupling between photons and an electronic band.

This tunability of an electronic structure should be one of the most unique points of GOS. By virtue of this property, GOS can play various roles in either electronic or photonic application, just by changing the crystallographic orientation of the Si substrate, as shown in Fig. 4. GOS using Si(111) as the substrate can serve as the channel for the FET of digital operation, particularly at the terahertz frequency. On the other hand, GOS using Si(100) and Si(110) as the substrates can serve as laser components for ultrahigh-speed optical communication. In the near future, we anticipate GOS-based electronic-photonic fusion integrated circuits; therefore, we are exerting effort to fabricate nanostructured GOS, owing to our collaborative work at BL17SU on spectroscopic low-energy electron microscopy (SPELEEM).

	On Si (111)	On Si (100)/Si (110)
Interface Structure	graphene / buffer layer / SiC / Si	graphene / SiC / Si
Stacking (Symmetry)	Bernal stack	Non-Bernal stack
Band Structure	• Band split • Gap open	• No band split • Gap close
Device Application	Logic FET	Laser

Multifunction LS1
- Low Power
- Rare-Metal Free

Fig. 4. Multifunctionality of GOS depending on the orientation of the substrate.

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