Realization of continuous Zachariasen carbon monolayer

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Rapid progress in two-dimensional (2D) crystalline materials has recently enabled a range of device possibilities. These possibilities may be further expanded through the development of advanced 2D glass materials. Zachariasen carbon monolayer, a novel amorphous 2D carbon allotrope, was successfully synthesized on germanium surface. The one-atom-thick continuous amorphous layer, in which the in-plane carbon network was fully sp²-hybridized, was achieved at high temperatures (>900°C) and a controlled growth rate. We verified that the charge carriers within the Zachariasen carbon monolayer are strongly localized to display Anderson insulating behavior and a large negative magnetoresistance. This new 2D glass also exhibited a unique ability as an atom-thick interface layer, allowing the deposition of an atomically flat dielectric film. It can be adopted in conventional semiconductor and display processing or used in the fabrication of flexible devices consisting of thin inorganic layers.

INTRODUCTION

Amorphous materials are widely playing essential roles in the electronics industry due to their superb electronic, mechanical, and thermal properties together with practical advantages, including low cost, uniformity, and excellent processability (1). For example, silicon dioxide (SiO₂) and other advanced oxides are the most important dielectric material used in memory nodes of Flash and DRAM, and gate oxide in logic devices. Liquid crystal display panels have been operated using amorphous silicon thin-film transistor (TFT). Moreover, amorphous metal oxide semiconductors (for example, IGZO) are becoming the replacement of crystalline Si for TFTs in organic light-emitting diode displays (2). Recent remarkable progress in two-dimensional (2D) materials has opened a range of possibilities for semiconducting and semimetallic device components. However, most of these important contributions have been seeking 2D crystalline resurrections of 3D materials. So far, 2D glass material, as a counterpart of 3D glasses above, has not yet been addressed adequately.

Glass materials were first defined by Zachariasen in the 1930s (3), and his requirements for a glass can be summarized as follows: (i) a glass should be a continuous and random network, and (ii) a glass should be formed from a specific form of atomic building blocks, which are also used in corresponding crystalline materials. In addition, (iii) a 2D glass should be a distinct atomic monolayer formed by strictly 2D arrangements of atoms. Recently, Shaikhutdinov and Freund (4) reported growth of ultrathin amorphous SiO₂ layer on metal surface, in which tetrahedral SiO₄ building units are randomly connected to form uniform bilayer structures. The amorphous SiO₂ bilayer meets Zachariasen’s requirements of an amorphous solid [requirements (i) and (ii)] but does not fulfill the atomic monolayer requirement of 2D glass [requirement (iii)]. On the other hand, a continuous 2D random network of sp²-hybridized carbons [Zachariasen carbon monolayer (Z-CM)] certainly satisfies all the requirements of Zachariasen 2D glass (5–7). Geim (8) first confirmed the existence of extended defects in a very small region with high-resolution transmission electron microscopy (HR-TEM), in which local rings consisted of pentagons, hexagons, and heptagons.

Because Z-CM is not present in nature, a variety of approaches have been experimentally tested to realize the 2D carbon glass (9–17). A possible synthetic approach is to lower the growth temperature during the chemical vapor deposition (CVD) growth process (9, 10). At the low reaction temperature near 600°C, the hydrocarbon precursor (for example, CH₄) is incompletely dehydrogenated, and the carbon adatoms do not diffuse with sufficient mobility on the substrate, thereby hindering the formation of a periodic arrangement. An atomically thin amorphous film can then form, in principle. However, a uniform one-atom-thick layer was not achieved on either catalytic (Ni) or non-catalytic (MgO) substrates (9, 10). The defective carbon films did not meet the atomic monolayer requirement [requirement (iii)] of a 2D Zachariasen glass. For another synthetic method, Turchanin et al. (15) converted an aromatic self-assembled monolayer to a highly disordered nanocrystalline graphene, in which more than 60% of the area was covered with crystalline nanopatches with 2- to 5-nm grain size (Lg), and the nanocrystalline patches were connected with amorphous carbon structure (16). However, the highly disordered graphene did not meet the continuous and random network requirement [requirement (i)] of a 2D Zachariasen glass.

An alternative approach would be the intentional generation of disorder on a crystalline graphene monolayer. Ion (or electron) beam exposure on crystalline graphene can mechanically break carbon bonds to form enormous structural defects and perturb the periodicity in the atomic arrangement (11–14). However, the bond breaking process inevitably generates large numbers of carbon vacancies (12, 14, 17). High irradiation doses are required to initiate the transition from a crystalline to an amorphous phase, and the resulting carbon density deficit can exceed 15% (17). These structural properties unfortunately do not satisfy the continuity requirement [requirement (i)].

Here, we have grown a one-atom-thick Z-CM on a Ge surface using conventional CVD processes. A Ge surface was used as the catalytic substrate for the growth of a continuous 2D carbon glass due to...
the following reasons. The lattice mismatch with graphene is large, which can readily hinder the formation of a periodic atomic arrangement. For example, the large lattice constant of Ge (5.65 Å) and the nearest neighbor atomic distance (3.995 Å) in a (111) surface yield a lattice mismatch with graphene of 62.4% (graphene has a lattice constant of 2.46 Å; see table S1) (18). Furthermore, germanium offers an extremely low carbon solubility (<10⁸ atoms/cm³), even at its melting temperature. Therefore, the growth process is restricted to the Ge surface and yields atomic monolayer (19). These chemical and physical properties of Ge, weak interaction with graphene and low carbon solubility, are also helpful for excluding unwanted disorder relaxation under our growth conditions (19, 20).

RESULTS

Wafer-scale synthesis of Z-CM

The islands of layered carbon grown on a Ge surface were divided into two distinguishable regions consisting of a bright center and a surrounding dark area (Fig. 1A). By tuning the growth conditions, such as H₂ partial pressure (P_{H₂}), we can control the size of the bright region of the layered carbon islands (Fig. 1, B and C, figs. S1 to S4). To minimize the external effects caused by oxidizing impurities in precursors (21), we conducted all experiments by using a purified gas condition (UHP grade, O₂ and H₂O < 1 ppb). Under the fast growth conditions in which P_{H₂} = 5 torr and the CH₄/H₂ ratio was 1:5, the entire wafer was covered only by the bright region. Decreasing the

Fig. 1. Islands of layered carbon grown on a Ge(100) surface and spectroscopic analysis of our Z-CM. (A) A typical scanning electron microscopy (SEM) image of layered carbon islands during the early stages of growth. Each carbon island is divided into two distinct areas: bright and dark. (B) The diameter of the bright carbon layer area depended on the H₂ partial pressure, at a constant ratio of the CH₄/H₂ inlet gases. (C) SEM images of the layered carbon islands under different P_{H₂} conditions at CH₄/H₂ = 1:100 after the growth times of 5, 5, 10, and 30 min, respectively. (D) Raman spectra of Z-CM and c-Gr at 514.5 nm. a.u., arbitrary units. (E) X-ray photoelectron spectroscopy (XPS) patterns (C 1s peak) obtained from Z-CM and c-Gr. The spectra were fitted to a single Doniach-Sunjic (D-S) model.
growth rate by increasing $P_{\text{H}_2}$ to 100 torr with a CH$_4$/H$_2$ ratio of 1:100 provided the dark carbon layer fully covering the wafer. The dark carbon layer was readily characterized to be a honeycomb-structured crystalline graphene via Raman analysis and HR-TEM image (fig. S5). However, the bright carbon layer showed amorphous behavior totally different from that of the dark one. The amorphous network might be thermodynamically less stable than the crystalline one, but the amorphous carbon layer was also quite stable in ambient conditions even at high temperature (300°C) (fig. S6).

To figure out whether it meets all the requirements of 2D Zachariasen glass, we carefully characterized the amorphous carbon layer using various analytical methods. For convenience, the bright carbon layer grown on the Ge substrate is called Z-CM below. The Raman spectrum of the Z-CM on a SiO$_2$/Si wafer showed its highly disordered nature (Fig. 1D). A sharp 2D band at 2800 cm$^{-1}$, attributed to the breathing modes of crystalline graphene, nearly disappeared in the amorphous 2D layer. The D mode ($A_{1g}$ type), which is forbidden in an ideal graphene, was highly activated by the structural randomness ($21, 22$). The G mode ($E_{2g}$ type) was significantly broadened by the presence of various different structures connecting the sp$^2$-hybridized carbon atoms ($22, 23$). The ratio of $I(D)/I(G)$ is usually considered a key measure for the amorphization in various carbon films, but it is still ambiguous. Ferrari and Robertson’s (23) three-stage model for phenomenologically interpreting disordered and amorphous carbon indicated that the amorphous carbon with the 20% of sp$^3$ centers has the $I(D)/I(G)$ ratio of ~0.2. More recently, Lucchese et al. (24) and Eckmann et al. (25) estimated that the minimum $I(D)/I(G)$ for ideal amorphous 2D network of sp$^2$-hybridized carbons (sometimes called amorphous graphene, $L_a$ ~ 0) is ca. 0.8 on the basis of e-beam (or plasma)–induced disordered graphene studies. According to their model, our Z-CM showing a $I(D)/I(G)$ ratio of ~1 is very close to the amorphous phase. Considering the experimental variation, it is hard to clearly discern between the amorphous and the nanocrystalline phase with 1- to 2-nm-sized nanograins from Raman spectrum. Other major parameters, position and bandwidth of G band, also show a large difference from Ferrari’s model, which indicates that the amount of sp$^3$ centers within our Z-CM is significantly lower than 20% (detailed interpretation of Raman analysis in note S1). The fraction of sp$^3$ carbon centers was also evaluated by analyzing the C 1s peak in the XPS profile (see note S2). Figure 1E shows that the C 1s peak of Z-CM was slightly broader than the corresponding crystalline peaks obtained from c-Gr or HOPG (crystalline graphene grown using the CVD method or highly ordered pyrolytic graphite, respectively); however, the asymmetric shapes of the peaks remained unchanged. The binding energy (284.5 eV) and the anisotropy factor (0.13) of the anisotropic peaks could be fit to a single D-S function describing sp$^2$-hybridized carbon, suggesting that other carbon components (especially sp$^3$-hybridized carbon located at 285 eV) were negligible (26). The fit parameters obtained from the D-S model (table S2) indicated that the Gaussian bandwidth corresponding to vibrational phonon and local configuration interactions contributed significantly to broadening because of the disordered character of the Z-CM (27). The Lorentzian terms corresponding to the photogenerated hole lifetimes in Z-CM and the crystalline graphene were indistinguishable (27). Therefore, the peak broadening was not attributed to the presence of different types (for example, sp$^3$) of carbon bonds but rather appeared to arise from the structural randomness. The Raman and XPS results show that the building block of Z-CM is sp$^2$-hybridized carbon [requirement (ii), building block].

HR-TEM images of our Z-CM grown on the Ge surface show that the carbon atoms are randomly arranged and do not form long-range periodicity in Fig. 2. The Fast Fourier Transform (FFT) pattern of the HR-TEM image (inset of Fig. 2A) also represented a continuous ring shape. In the Raman analysis above, the question of whether our Z-CM is a perfect amorphous phase or a nanocrystalline phase with very small nanograins (<3 nm) still remained. For this reason, we closely examined HR-TEM images and their FFT patterns in local areas (2.5 × 2.5 nm$^2$), which were selected in contamination-free regions,

Fig. 2. High-resolution transmission electron microscopy (HR-TEM) of Z-CM. (A) HR-TEM images of the bright area (Z-CM) of Fig. 1A. The inset shows its FFT pattern. (B) Fivefold enlarged image of white box in (A). The inset shows its FFT pattern. (C) FFT spectrum of the inset of (A) and Gaussian distribution of ring pattern obtained by fitting the spectrum.
but a nanocrystalline domain larger than 1 nm was not found (Fig. 2B and figs. S7 to S11). The broadening of a ring pattern is also a typical phenomenon in highly disordered (or defective) graphene. It occurs in both radial and azimuthal directions. According to the study by Eder et al. (17) on disordered graphene, the radial broadening was saturated to ca. 0.4 to 0.6 (1/nm) even at an ideal 2D amorphous structure. In the FFT pattern of our Z-CM, radial broadening was estimated to be 0.45 to 0.51 (1/nm), which agreed with Eder’s experiments (Fig. 2C). Small, hexagonally arranged carbon clusters (La < 1 nm) may be formed in our Z-CM grown by the CVD method, but the proportion will be negligibly small. Notably, some degree of order intermediate between a liquid and a crystal exists in many glass structures, according to Martin et al.’s understanding of the amorphous phase (28).

For the monolayer requirement, we verified the atomic thickness of Z-CM using atomic force microscopy (AFM) and cross-sectional TEM imaging methods. Figure 3A shows a typical optical image of a sample with vertically stacked Z-CM and c-Gr layers. We also took electrostatic force microscopy (EFM) images of various stacked structures. The monolayer and bilayer can be distinguished on the basis of the EFM images (see the insets of Fig. 3, B and D). The monolayer was darker in color than the bilayer in the EFM images. The thickness of the crystalline graphene, determined by the steps between the Z-CM and c-Gr/Z-CM layers (Fig. 3B and C, and fig. S12), was 3.5 Å and agreed well with results reported previously (29). On the other hand, the thickness of the Z-CM layer, measured from the steps between the c-Gr and c-Gr/Z-CM layers (Fig. 3D and fig. S13), was slightly thicker (4.3 Å) than the corresponding step of the c-Gr monolayer. We attribute this difference to the puckered nature of Z-CM produced by odd-membered rings (for example, pentagonal and heptagonal rings) (4). A cross-sectional TEM image of the as-grown Z-CM on a Ge wafer further confirmed that a one-atom-thick layer was formed (Fig. 3E). The large-area uniformity of the Z-CM layer was apparent in the Raman mapping (60 × 70 μm²) of the G band intensity (Fig. 3F). All the results of Figs. 1 to 3 confirmed that Z-CM is the one-atom-thick 2D glass film that satisfies all requirements proposed by Zachariasen.

Electronic property

The disordered structure of the Z-CM induces electron backscattering corresponding to Anderson insulating behavior (30–32). Figure 4A shows that the electrical resistance of Z-CM was two orders of magnitude higher than that measured in c-Gr. Unlike crystalline graphene, thermal treatment (at 300°C for 2 hours under a H2/Ar environment) could not reestablish the charge neutrality point of the Z-CM field-effect transistor (FET) device at a zero gate bias. This is due to the easy chemical adsorption characteristic of Z-CM (fig. S14). The temperature dependence of Z-CM was obtained using four-point measurements. The resistance in Fig. 4B significantly increased across the low-temperature regime (below 50 K) and was well fitted to Mott’s 2D variable-range hopping (VRH) relation

\[ \rho(T) \sim \exp \left( \frac{T_0}{T} \right)^{1/3} \]

indicating strongly localized charge carriers and Anderson insulating behavior of Z-CM (33). The strongly localized carriers displayed a large negative magnetoresistance (MR) effect below 100 K (Fig. 4C) upon application of the perpendicular magnetic field to the Z-CM.
resistance, based on four-probe measurements collected from Z-CM. The red line shows the fitting results to Mott's VRH model \( \rho(T) \sim \exp \left( \frac{T}{T_D} \right) ^{3/2} \). (C) The large negative MR behavior of Z-CM at low temperatures below 100 K. \( R_{xx} \) is a longitudinal sheet resistance.

FET device. The MR effect increased markedly as the temperature decreased, reaching the level of the giant MR (~30%) at 2 T. The negative MR curve followed Zhao's relation \( \rho(B) \sim \exp(-B^{1/2}) \), which is based on the 2D VRH theory, as shown in fig. S15 (34, 35).

Previous studies have reported that disordered 2D carbon materials tend to undergo a strong-to-weak localization (SL and WL, respectively) phase transition as the charge carrier concentration increases (30–32). The boundary between the SL and WL is determined by two characteristic lengths: the localization length \( \xi_L \) and the phase coherence length \( L_C \). Zhou's experimental results predicted that \( \xi_D \) can be reduced down to 0.1 nm, but \( L_C \) becomes saturated around 3 nm as the amorphicity of graphene increases. In the nanocrystalline regime, the two characteristic lengths become comparable, and the WL behavior emerges at a high carrier concentration (36). On the other hand, because Z-CM includes a degree of disorder that approaches the amorphous phase, \( \xi_D \) is much shorter than \( L_C \) and satisfies the conditions for SL, even at high concentrations.

**Unique functionality for device application**

Another important property of our Z-CM is a unique function as atom-thick interfacial layer for the deposition of heterolayers. Crystalline graphene is known to have inert surface and usually requires an additional seed formation process for the subsequent material deposition (37), except in a few special cases such as growing lattice matched layers (38). In our study, a continuous Al2O3 layer could not be obtained on a c-Gr control (Fig. 5A). On the other hand, because of the inhomogeneous distribution of charge density originated from random sp3 carbon bonding (Fig. 2), the Z-CM can serve as a continuous one-atom-thick seed layer. Atomic layer deposition (ALD) of a thin layer (12 nm) of Al2O3 onto Z-CM without any treatment, such as O2 plasma, produced an ultrasmooth, flat continuous layer (Fig. 5, B and C). By introducing Z-CM interlayer, we could fabricate metal oxide semiconductor (MOS) capacitor on pristine hydrogenated Ge(110) surface. Figure 5D depicts the normalized capacitance-voltage \( C-V \) characteristics of the MOS capacitors Au (20 nm)/Ti (5 nm)/Al2O3 (15 nm)/Z-CM/Ge(110). It was found that the control MOS capacitor without the Z-CM interlayer exhibited stretched-out \( C-V \) behavior and the large hysteresis loop, indicating that a large number of charge traps exist at the interface of Al2O3/Ge (39–41). These problems are attributed to the easy formation of germanium oxide at high-k/Ge interface, which have been considered one of the major obstacles for using Ge in semiconductor devices (39, 40). In contrast, the MOS capacitor with the Z-CM interlayer showed a significant improvement in \( C-V \) characteristics. The hysteresis was almost removed, and the \( C-V \) slope also became steeper, proving that oxidation of Ge at the Al2O3/Ge interface was effectively suppressed. Another advantage of introducing the Z-CM interlayer is that hydrogenated Ge surface is naturally formed during CVD growth of Z-CM and is effectively passivated by the Z-CM overlayer from atmospheric oxygen. Although various approaches addressing the high-k/Ge interface and their significant advances have been demonstrated, more cost-effective and reliable process technologies are still required. This Z-CM interlayer could be a solution to enhance the oxide/Ge interface properties and Ge MOS capacitor process.

Z-CM can be also used as an atomic-thick seed layer for fabricating ultrathin silicon layer (thickness, <1 μm). Relatively weak interactions between the synthesized Z-CM and the underlying hydrogen-terminated Ge surface (19) enabled facile mechanical exfoliation of the Z-CM layer together with the upper silicon layer. The fabrication and transfer processes for the ultrathin silicon devices are briefly summarized in fig. S16. Ultrathin silicon patterned using universal photolithography methods could be released using polydimethylsiloxane stamp from the mother substrate (Ge wafer) and successfully transferred into the SiO2 wafer or flexible substrates (Fig. 5E). We note that highly defective graphene layers and other ultrathin buffer layers have also shown impressive 2D seeding behaviors (see table S3). Currently, direct comparison of Z-CM with the previously reported seeding layers is not of great importance because they have been evaluated in a variety of experimental conditions, and especially, their performances as seeding layer were demonstrated in local area or a unit device. However, considering future demands for highly integrated devices with sub-10-nm feature size, amorphous phase and single atomic thickness of Z-CM must be attractive for forming ultrathin dielectric layer with high uniformity. Direct growth of Z-CM on semiconductor Ge with complementary MOS compatible process will be another merit.

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Fig. 4. The electronic properties of Z-CM. (A) Sheet resistance curves obtained from Z-CM and c-Gr FETs as a function of the gate bias, measured under high-vacuum conditions (1 × 10^-8 torr). The inset shows the results measured from a graphene FET prepared with a Hall bar structure. (B) Low-temperature dependence of the sheet resistance, based on four-probe measurements collected from Z-CM. The red line shows the fitting results to Mott's VRH model \( \rho(T) \sim \exp \left( \frac{T}{T_D} \right) ^{3/2} \). (C) The large negative MR behavior of Z-CM at low temperatures below 100 K. \( R_{xx} \) is a longitudinal sheet resistance.
DISCUSSION
We synthesized Z-CM, uniform one-atom-thick amorphous network of sp²-hybridized carbons, and found its extraordinary properties totally different from conventional crystalline graphene. For example, Z-CM has shown Anderson insulating behavior, which is a fascinating feature applicable to magnetic devices, although it may not reach the insulating performance of the SiO₂ bilayer with intrinsic large band gap ($4\sigma$). It has also shown reasonably good chemical and thermal stability, enabling its practical applications. A couple of interesting behaviors of Z-CM demonstrated here, including seeding layer performance for upside layer formation as well as passivation layer for downside layer, show a very small part of its real potential. We anticipate that our study of Z-CM will lead to active application of 2D glass materials in practical fields like other amorphous bulk materials such as a-Si and a-SiO₂ in current electronics technology.

MATERIALS AND METHODS

Growth of Z-CM on a Ge surface
A Ge wafer or Ge-deposited Si wafer was cleaned using the standard RCA method followed by oxygen plasma treatment to remove organic residues. The Ge substrate was then dipped into a 10% HF solution to remove native oxide groups. The H-terminated Ge substrate was immediately loaded into a low-pressure CVD (LPCVD) chamber (SETS Co.) and the chamber was evacuated until ~5 x 10⁻⁶ torr. To avoid internal and external contamination issues, we installed gas purifiers for all precursors (Entegris GateKeeper Series), and we deposited a fresh epitaxial Ge layer on the Ge substrate and reacting zone by flowing 40 sccm of GeH₄ gas (10% in hydrogen, 99.999%, ultrahigh purity grade, O₂ and H₂O < 1 ppb) at 500°C for 20 min with 5 to 30 torr. Z-CM was synthesized using a mixture of CH₄ and H₂ gases (99.999%, ultrahigh purity grade, O₂ and H₂O < 1 ppb) introduced into the chamber at 900° to 930°C. Before Z-CM synthesis, the chamber atmosphere was changed from H₂ to Ar to enhance the graphene growth rate. During the growth process, the total pressure was maintained at 1 to 10 torr over 5 to 30 min. Last, the as-grown substrate was rapidly cooled to room temperature under a hydrogen atmosphere to avoid recrystallization of Z-CM on Ge (42).

Growth of an ultrathin Al₂O₃ layer grown on a Z-CM surface
An ultrathin Al₂O₃ layer was deposited on a Z-CM surface using ALD without any pretreatments. The Ge wafer (or Ge-deposited Si wafer) supporting the as-grown Z-CM was loaded into the ALD reactor and vacuumed down below 5 x 10⁻⁶ torr. Substrate temperature was set to 200°C. Before Al₂O₃ deposition, a sample was held under vacuum over 20 min and purged with Ar gas for 3 min. Trimethylaluminum (TMA) and H₂O were subsequently exposed in the reactor: dose TMA for 0.3 s, dose Ar for 10 s, dose H₂O for 0.3 s, and then dose Ar for 10 s. This was one cycle of Al₂O₃ deposition. This cycle was repeated 50 to 120 times.

Fig. 5. Dielectric layer (Al₂O₃) deposited on Z-CM and ultrathin silicon layer fabrication using Z-CM. (A) SEM image of Al₂O₃ deposited onto c-Gr via ALD. The inset shows an SEM image of Al₂O₃ uniformly deposited onto Z-CM using the same method. (B) AFM image of Al₂O₃ deposited on Z-CM where surface roughness was 2.17 Å root mean square (RMS). (C) Cross-sectional TEM image of (B). (D) C-V characteristics measured at 100 kHz from Al₂O₃/Z-CM/Ge and Al₂O₃/Ge capacitors. The C-V curves were normalized with respect to their maximum accumulation capacitances. (E) Patterning and transferring of ultrathin amorphous silicon layer deposited on Z-CM/Ge.

Growth of an ultrathin silicon layer on a Z-CM surface

An ultrathin silicon layer was grown on a Z-CM surface using conventional LPCVD methods, similar to the Al2O3 layer deposition explained above. The Z-CM as-grown on the Ge wafer (or Ge-deposited Si wafer) was loaded into another LPCVD chamber. If the silicon layer had been deposited using a silicon precursor (for example, SiH₄) in the graphene growth chamber, then cross-contamination of the chamber could not have been avoided. The silicon layer was deposited by introducing 50 sccm of SiH₄ gas (5% in He) (125 mtorr) with H₂ (50 sccm) at 600°C for 10 to 60 min. The total pressure was maintained at 5 torr. The silicon layer could be in situ–doped by mixing small amounts of the PH₃ gas (n-type dopant) or B₂H₆ (p-type dopant) gas together with the SiH₄ gas (fig. S17).

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/3/2/e1601821/DC1

Note S1. Raman analysis.

Table S1. Lattice mismatch of typical catalytic substrates for graphene growth.

Table S2. Fitting parameters from the D-S model.

Table S3. Summary of previous studies on ultrathin dielectric layer deposition on graphene.

Table S4. Lattice mismatch of typical catalytic substrates for graphene growth.

Note S2. XPS analysis.

Table S5. List of references.

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