Evidence for a pressure-induced phase transition of few-layer graphene to 2D diamond

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We unveil the diamondization mechanism of few-layer graphene compressed in the presence of water, providing robust evidence for the pressure-induced formation of 2D diamond. High-pressure Raman spectroscopy provides evidence of a phase transition occurring at ≈ 6 GPa for bi-layer graphene, and in the range of 4–7 GPa for 5-layer graphene and graphite. The pressure-induced phase is partially transparent and indents the silicon substrate. A nucleation-propagation model is proposed as the diamondization mechanism, allowing for the formation of diamondene, a 2D ferromagnetic semiconductor. High-pressure x-ray diffraction on a graphene powder indicates the formation of hexagonal diamond, consistent with the bulk limit of eclipsed-conformed diamondene.

The search for a stable 2D diamond has gathered recent interest due to the possibility of combining diamond’s distinguished properties, such as superior hardness [11] and heat conduction [2], to exotic new properties that may arise from the reduced dimensionality. Its existence was first proposed over a decade ago [3] and different structures have been theoretically proposed ever since [4,7]. In most structures, stability is achieved by surface functionalization at the top and bottom surfaces [3,6,7], sometimes called diamane [3] for bilayer, and diamondoids for thicker layers [8]. Diamondene [9], another proposed 2D-diamond structure, greatly differs from those, being covalently bonded to chemical groups only at the top surface while the bottom exhibits a periodic array of dangling bonds. This structure is thermodynamically stable and, as long as the substrate is chemically inert, the dangling bonds will be preserved. These unpaired electrons generate magnetism in diamondene and their periodic distribution gives rise to two spin-polarized bands, making it an ideal platform material for spintronics [5,9].

In this letter, we investigate the formation of diamondene via high-pressure experiments, using a diamond anvil cell (DAC) and we provide structural information through high-pressure x-ray diffraction (XRD). We use water as the pressure transmitting medium (PTM) due to its importance in facilitating this phase transition [5,9]. High-pressure Raman spectroscopy provides evidence of a phase transition occurring at ≈ 6 GPa for bi-layer graphene, and in the range of 4–7 GPa for 5-layer graphene and graphite. The pressure-induced phase is partially transparent and indents the silicon substrate, indicative of extreme hardness. We combine experimental data with Molecular Dynamics (MD)/Density Functional Theory (DFT) simulations to propose a full diamondization mechanism in compressed few-layer graphene, providing robust evidence for the pressure-induced formation of 2D diamond. We show that the use of water PTM allows for a gradual top-bottom diamondization process, consistent with the formation of diamondene.

High-pressure Raman experiments were performed on mechanically exfoliated graphene samples transferred onto 25-µm-thick Si substrates with a 300 nm-thick SiO2 layer (see details in Supplementary Information). Two samples were prepared using this method: a bi-layer graphene, shown in Fig. S1(c), and a graphite piece sitting next to a five-layer graphene, both shown in Fig. S1(d). The samples were compressed in the DAC using water as the PTM, and all Raman spectra were acquired using a 532 nm excitation laser. The hydrostatic limit of water is discussed in Supplementary Information. Figures 1(a–c) show selected Raman spectra of the compressed bilayer graphene, five-layer graphene and graphite, respectively, measured at different pressures featuring the first-order Raman-allowed G band (≈ 1580 cm–1 in ambient pressure), and the two-phonon Raman 2D band (≈ 2700 cm–1) [10].

Figures 1(d–f) show the plot of the pressure-evolution of the G and 2D band frequencies subtracted from their frequencies measured in ambient conditions (ΔωG and Δω2D, respectively). The data were extracted from the Raman spectra of bilayer graphene, five-layer graphene and graphite respectively. Figures 1(d–f) further show the evolution of the full-width at half maximum of the G band, ΔΓG. At low pressures, we observe the expected strain-induced phonon hardening [11], by shifts in the G and 2D bands [12] to higher frequencies without significant changes in ΓG [13]. However,
FIG. 1. Phase transition evidence obtained from Raman spectroscopy. (a–c) Selected Raman spectra at different pressures for (a) bi-layer graphene, (b) five-layer graphene and (c) graphite. G and 2D bands are indicated with * and ↓, respectively. (d–f) Plots of the G (2D) band frequency and full width at half maximum subtracted from their values at initial pressure, ∆ω_G (∆ω_{2D}), and ∆Γ_G, respectively, as a function of pressure for (d) bi-layer graphene, (e) five-layer graphene and (f) graphite. The blue-dashed lines indicate the critical pressures of the phase transition. (g–i) Plots of the intensity ratio between the 2D and G band (I_{2D}/I_G) for (g) bi-layer graphene, (h) five-layer graphene and (i) graphite.

this trend changes at ≈6 GPa for bilayer graphene and at ≈4 and ≈7 GPa for both five-layer graphene and graphite. These critical pressures are evidenced by the blue dashed lines in Figs. 1(d–f). The most remarkable change is an abrupt increase in Γ_G, which is considered a signature of structural phase transitions in compressed graphite [14–16]. For both 5-layer [Figs. 1(b,e)] and graphite [Figs. 1(c,f)], Γ_G suddenly broadens around 4 GPa indicating the onset of a phase transition, and remains relatively constant above 7 GPa, indicating a completed transition.

The occurrence of a phase transition is further bolstered by the concomitant abrupt intensity suppression of the 2D band, as observed in the Raman spectra shown in Figs. 1(a–c) and systematically depicted in the plots of the ratio between the 2D and G bands intensities, I_{2D}/I_G, as a function of pressure, shown in Figs. 1(g–i) for the bilayer graphene, five-layer graphene and graphite, respectively. Since the 2D band originates from double-resonance mechanisms [10], its intensity is highly sensitive to changes in the electronic structure [17]. Changes in the slope of G band frequency with pressure provide additional evidence of phase transition (see discussion in Supplementary Information). The totality of information extracted from the Raman spectroscopy data presented in Fig. 1 provides strong indications that a sharp structural phase transition occurs at 6.3 GPa for the bilayer graphene, whereas for both 5-layer graphene and graphite, a transition occurs over the range from ≈4 GPa to ≈7 GPa.

Consistent with the Raman spectroscopic signatures of a pressure-induced phase transition, changes in the optical properties of the 5–layer graphene and graphite flakes were also observed in the 4–7 GPa range. Figure S2 shows optical images of the sample inside the DAC at different pressures. Starting at 4.5 GPa, a series of color changes occur in the graphite flake, with the formation of a yellow region which spreads over the flake with increasing pressure, then gradually turns to a color similar to the substrate – an indication of increasing transparency. The 5–layer graphene and graphite flakes can no longer be seen at 6.4 GPa and 7.1 GPa, respectively. Indeed, after 7.1 GPa, the Raman peak coming from the SiO_2/Si substrate (≈1520 cm^{-1}), which is
FIG. 2. Evidence of pressure-induced transparency and formation of a hard phase. (a) Raman spectra of graphite at different pressures. Notice that, from 7.1 GPa, the Raman peak from the silicon substrate appears at approximately 550 cm$^{-1}$. (b) Plot of the ratio between the intensity of the Si Raman peak measured on graphite ($I_{Si,Gt}$) and measured on bare silicon substrate ($I_{Si}$), as a function of pressure. As the graphic shows, the ratio becomes $\approx 0.25$ for pressures above 7.1 GPa, indicating that the graphite flake becomes partially transparent.

Upon phase transition, from the optical images we observed cracks on the substrate along directions defined by the flake edges (Figure S3), indicating the formation of a hard phase. To confirm this feature (see discussion in Supplementary Information), we compressed a sample containing a 4–layer graphene and a graphite flake on SiO$_2$/Si in a DAC, up to 8 GPa using water PTM. The AFM topographical images of the recovered sample clearly show indentation marks on the SiO$_2$, following the shape of both graphite [Fig. 2(c)] and 4–layer samples (Fig. S4). Furthermore, the Raman spectra of the recovered samples are remarkably similar to those of hydrogenated graphene [19], being consistent with a partial functionalization of graphene after compression (see Fig. S5 and related discussion in Supplementary Information). In addition, the symmetric shape of the 2D band indicates the occurrence of decoupling between graphene layers [10].

To obtain information on the crystal structure of the new phase, we have performed high-pressure XRD on a graphene powder using water PTM. A graphene powder was used instead of a single few-layer graphene crystal, since the latter...
The rise of a peak with similar features has been reported in room-temperature compressed graphite upon decompression and assigned as 002 of hexagonal diamond (HD) \[18\]. As shown in Fig. 3(b), there is a good agreement for the two strongest peaks HD002 and HD100. Other HD peaks could not be identified likely due to a combination of low intensity, unfavorable spatial orientation of flakes that meet the Bragg condition (similar explanation holds for the absence of some of graphene peaks) and overlap with ICE VII or graphene peaks, as can be seen in Fig. S14. In fact, it is possible to see the rise of the HD002 peak at approximately 14 GPa [Fig. 3(c)]

To interpret the experimental results, we carried out MD simulations to dynamically follow the restructuring process at a given temperature and DFT calculations to study the stacking order of the rehybridized structure and the interplay between structural and electronic properties. Both theoretical schemes assume a starting model of AB–stacked five–layer graphene whose top surface is covered with either –OH or –H groups. These are assumed to originate from water molecules in reactions under high pressures at the surface of graphene \[5\] (see details in Supplementary Information), and play a fundamental role in both the rehybridization and stabilization processes \[20\].

Figure S8 presents the profiles of three snapshots with increasing pressure obtained from MD simulations, clearly showing the evolution of sp\(^3\) bond formation. Under compression, the phase transition starts with the diamondization of the first two layers, giving rise to diamondene. This happens in two steps, the first being nucleation: the formation of random chemical bonds between the two layers due to their closer proximity, allowing for the sp\(^2–sp\(^3\) rehybridization of the carbon atoms. Such a process is greatly facilitated by the presence of –H groups on the top surface. Once these chemical bonds are formed, they trigger the sp\(^3\) conversion across the first two layers through a cooperative phenomenon \[21\] in which each tetrahedral bond favors the formation of a neighboring sp\(^3\) bond. As a result, the diamondene structure is formed in this so-called horizontal propagation (see details in Supplementary Information). For the formation of covalent bonds between diamondene and the graphene layer underneath to occur, the pressure needs to be further increased since the distance between the layers is too large to allow for the rehybridization. In this process, called vertical propagation, the third graphene layer displaces horizontally resulting in a transformation from an ABA to an ABBB trilayer stacking configuration.

Figs. 4(a,b) show the optimized geometries for two configurations with increasing applied force, obtained from DFT calculations. In agreement with the MD simulations, the diamondene structure is initially formed with the underlying three-layer graphene protecting the array of dangling bonds [Fig. 4(a)]. Interestingly, a similar structure has been recently reported from hot-filament process of few-layer graphene's large compressibility along c-axis, and its intensity remains practically constant until approximately 5 GPa, after which, it starts to decrease, becoming undetectable after 15 GPa. Such decrease can be assigned to the onset of the phase transition, in agreement with the onset pressures observed in the Raman experiments (see discussion in the Supplementary Information). Upon decompression [diffractograms shown in Fig. 3(c)] a new peak is resolvable between G101 and G100. The rise of a peak with similar features has been reported in room-temperature compressed graphite upon decompression and assigned as 002 of hexagonal diamond (HD) \[18\]. As shown in Fig. 3(b), there is a good agreement for the two strongest peaks HD002 and HD100. Other HD peaks could not be identified likely due to a combination of low intensity, unfavorable spatial orientation of flakes that meet the Bragg condition (similar explanation holds for the absence of some of graphene peaks) and overlap with ICE VII or graphene peaks, as can be seen in Fig. S14. In fact, it is possible to see the rise of the HD002 peak at approximately 14 GPa [Fig. 3(c)].

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graphene [22]. Next, by increasing the applied forces, the diamondization process propagates through the other layers, resulting in a completely rehybridized five-layer geometry characterized by an ABBBB stacking, as shown in Fig. 4(b). In the BB part of that stacking, the interlayer bonds are in an eclipsed conformation. The electronic band structure corresponding to the ABBBB stacking, shown in Fig. 4(c), preserves the main features of the diamondene – localized states at the dangling bonds leading to a pair of spin-polarized bands. It is important to mention that different rehybridized geometries are also possible (see discussion in Supplementary Information).

This nucleation-propagation model explains several features of the experiment, for instance, the XRD diffractograms. Since there is no indication of significant water content inside the graphene powder (see discussion in Supplementary Information), it is reasonable to propose a top-bottom diamondization process as shown in Figs. 4(a,b), starting at the surface (in contact with water), and propagating toward the powder’s core. As the crystal grows, in the bulk limit, one should expect a BB stacked configuration [Fig. 4(b)], which is precisely the crystal structure of hexagonal diamond. It is also in the bulk limit where one should expect a detectable XRD signal, with intensity strong enough to overcome the broad graphene peaks. Moreover, the fact that the HD002 peak rises as the G002 graphene peak vanishes is consistent with the nucleation-propagation model, in which diamondene and graphene share the same c-axis.

The optical changes and the asymmetric shape of the G band for both 5-layer graphene and graphite after the onset of the phase transition can also be explained by our model. The appearance of a yellow color over the graphite flake shown in Fig. S2 is caused by the diamondization of the first top layers. As the pressure increases, the vertical propagation evolves and the diamondization extends to deeper layers, inducing further transparency of the flake. Also, starting from 4 GPa, the G band exhibits an asymmetric shape, which is more pronounced for graphite [Figs. 1(b,c)]. This could be caused by the coexisting pristine and hybridized phases during the vertical propagation [Fig. 4(a)], with distinct G band frequencies of narrowed and broadened bandwidth, respectively. This feature was confirmed and other aspects of the model were further investigated in a high-pressure Raman experiment comparing graphite and a graphene powder (see discussion in Supplementary Information). The nucleation-propagation model can explain the shape of the 2D band of the recovered samples post-compression. As observed from the Raman spectra obtained from the recovered samples, shown in Fig. S5, its symmetrical shape indicates that an overall loss of stacking order occurred in those flakes, consistent with a lateral sliding between layers during compression, in agreement with the vertical propagation mechanism.

Most previous experimental indications of the existence of a 2D diamond have been obtained either by tip-compression [5][7][23], hydrostatic compression [9][24] or hydrogenation of few-layer graphene [8][25]. Even though these works promoted significant advances in the field, they did not provide structural information about the new phase. Recent preprints investigate the formation of diamondene [20] and diamanoﬁds [22] via chemical-functionalization routes, reporting structural information through transmission electron microscopy and low-energy electron diffraction, respectively. Previous local high-pressure studies using atomic force microscopy (AFM) tips, reported a band gap opening in compressed bi- and multi-layer graphene [5], and the formation of an ultra-hard phase in two-layer graphene on SiC [7][23]. However, these works [5][7][23] showed different layer-dependence for the occurrence of the phase transition, exemplifying that the mechanism is not yet understood and that different conditions could lead to different structures.

Our combined experimental and theoretical analysis of the diamondization process from 2D to bulk sheds light on the long-standing problem on the formation of hexagonal diamond and other pressure-induced phases in cold compressed graphite [15][27][29]. Robust structural evidence of the pressure-induced formation of a 2D diamond was presented with a detailed description of the diamondization process. Raman spectroscopy data identify phase transition evidence and critical pressures for two- and five-layer graphene and graphite. Optical images and Raman spectroscopy on compressed graphite indicate a gradual increase of transparency due to a gradual top-bottom diamondization process. This gradual top-bottom diamondization process gives a hint of diamondene formation, that is, diamondized layers forming on the surface with underlying graphene layers protecting the dangling bonds. High-pressure XRD data provides structural information of the diamondization process in 2D flakes. The theoretical model describes in detail the diamondization process in the presence of passivation groups on the top layer as a gradual top-bottom process, which starts with the diamondization of the first two layers and then propagates along c axis to the bottom. Evidence of functionalization from the recovered samples point that a stable 2D diamond could be obtained through further tuning of the synthesis parameters, e.g. by increasing temperature, while to obtain diamondene, it is a matter of substrate choice. Potential applications of diamondene are in 2D magnetism, gate-tunable spin-polarized currents, and spin-polarized photocurrents, with broad impact in both fundamental research and practical applications.

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