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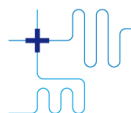
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ABSTRACT

Defect engineering is pivotal for tailoring graphene properties, yet conventional analysis often overlooks global structural connectivity—specifically, interconnection of individual carbon polygons to form extended defect clusters. Here, we investigated the structural dynamics of defects in metal-doped graphene and electron-irradiated graphene using *in situ* transmission electron microscopy (TEM operating at 80 kV below the theoretical knock-on threshold) combined with persistent homology (PH)-based analysis. PH is a quantitative and multi-scale analytical framework, describing the quantity and scale of k -dimensional holes. We have established an analytical strategy to investigate both the local and global structures of defects in graphene. In this study, we built upon the PH-based strategy by extending to include a dopant detection process for metal-doped graphene and then applied it to *in situ* TEM images in order to quantitatively study the effects of metal doping on defect cluster dynamics. The *in situ* observation demonstrated that the dopants generated defects at their vicinity and that Cr doping induced significantly faster structural changes compared to Cu doping under electron irradiation at the same dose rate. Furthermore, PH-based analysis quantitatively showed that Cr-doped graphene maintains higher defect connectivity of local polygon units and exhibits larger temporal fluctuations of the connectivity than electron-irradiated graphene, even at similar ratio of non-hexagonal polygons. Our *in situ* observation and analysis suggested that Cr atoms could be one of the key factors contributing to the formation of extended defects beyond the scale of the minimal carbon polygon units. Thus, analysis of polygon connectivity is essential for capturing the complex dynamics of disordered systems.

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Single-layered graphene, a two-dimensional monatomic sheet of carbon, has attracted great attention owing to its exceptional properties, including ultra-high electron mobility,¹ extraordinary mechanical strength,² and distinct electronic band structure.^{3,4} Precise modulation of these properties is important for fully realizing the potential of graphene in next-generation electronics, spintronics, and catalysis. This forms the foundation for defect engineering—a critical methodology involving the controlled introduction of structural imperfections, such as vacancies,⁵ and extended defects,^{6,7} to tailor the functionalities of graphene. This field encompasses both “bottom-up” strategies⁸ and “top-down” modification techniques.^{9,10}

In situ transmission electron microscopy (TEM) is a unique and powerful top-down approach.¹¹ Unlike bulk characterization techniques, such as x-ray photoelectron spectroscopy¹² and Raman

spectroscopy,¹³ TEM allows local structural analysis at the atomic level, making it an indispensable tool for characterizing graphene with structural imperfections. TEM also enables the dynamic induction, migration, or repair of graphene defects with atomic-level precision by controlling the electron beam parameters, such as the acceleration voltage, dose rate, and total dose. An important advantage of TEM is its simultaneous, real-time atomic-resolution imaging. Defect generation by TEM is mainly implemented via two modes: (1) direct displacement (knock-on) of carbon atoms by the electron beam irradiation^{14,15} and (2) interaction between the electron irradiation and pre-doped atoms.^{16–18}

The displacement of carbon atoms by electron irradiation is highly dependent on the acceleration voltage, and damage occurs when the energy exceeds the material-intrinsic knock-on threshold of

86 kV for graphene.¹⁹ Consequently, an acceleration voltage below the knock-on threshold is frequently used for general graphene observation. Studies on the interaction between metal dopants and electron irradiation have provided crucial insights into the stability and bonding of dopants within the graphene lattice. These studies have mainly focused on the local structure surrounding the metal atoms, such as coordination numbers and bond lengths of nearest-neighbor (NN) and next-nearest-neighbor (NNN) atoms.^{20,21} However, such a strictly local focus overlooks the broader global structural changes, and defect engineering requires a comprehensive, multi-scale understanding. For instance, it has been reported that self-doping of charge carriers can be achieved by controlling the connectivity of the carbon polygons to create line defects composed of octagons and pentagons,²² and that such line defects induce anisotropy in charge transport.²³ These studies underscore the importance of analyses that incorporate the connectivity of individual carbon polygons, the so-called defect clusters.

Here, to address the structural changes beyond the NNN scale, we used persistent homology (PH), a mathematical framework within topological data analysis, as a multi-scale analytical method. PH represents data through k -dimensional holes, providing information about their quantity and scale.²⁴ PH is becoming a leading method for capturing hierarchical structures, such as the short- and medium-range order in disordered materials such as amorphous solids and glasses.^{25,26} In recent years, image-based machine learning in materials informatics has mainly relied on convolutional neural networks (CNNs), which are inherently supervised and operate as black-box models. In contrast, PH is an unsupervised, white-box approach that does not require training data.²⁷ This enables direct chemical and physical discussion based on the analysis outputs. Unlike CNNs, which demand extensive training data, PH remains robust and applicable even when dealing with sparse experimental results or entirely new, unseen datasets.²⁸ We previously developed a PH-based strategy to extract both local information on individual carbon polygons and global information on clusters formed by these polygons from high-resolution TEM (HR-TEM) images of graphene.²⁹ In this study, we applied the PH-based analytical framework to graphene containing two types of metal dopants (copper and chromium) to investigate the defect generation and dynamics under electron irradiation from both local and global structural perspectives.

Commercially available single-layer graphene (Trivial Transfer Graphene, ACS Materials, USA) was transferred onto molybdenum-supported carbon films (Quantifoil, Quantifoil Micro Tools GmbH, Germany). To clean the graphene surface, the samples were annealed at 300 °C for 2 h under a high vacuum of 10^{-6} Pa. Metal atoms were then deposited with an Ar ion beam etching system (Gatan PECS, USA). Copper (Cu) and chromium (Cr) were selected as dopants based on previous reports indicating their ability to induce defect generation extending beyond the NNN scale under electron irradiation.^{30,31} For both metals, the ion beam energy was set to 10 keV with a deposition time of 1 s. Post-deposition annealing for decontamination was performed within the TEM column (approximately 10^{-6} Pa) at 600 °C for 6 h for the Cu-doped samples or at 450 °C for 3 h for the Cr-doped samples. Following annealing, the samples were observed *in situ* without exposure to air.

TEM observations were performed using an aberration-corrected transmission electron microscope (JEM-ARM200F, JEOL,

Japan). The metal-doped samples were observed at an acceleration voltage of 80 kV, with a dose rate of $2.3 \times 10^6 \text{ e}^- \text{ nm}^{-2} \text{ s}^{-1}$, while maintaining the TEM column pressure at 8.0×10^{-6} Pa. In addition, to clarify the influence of metal dopants on structural dynamics, an undoped graphene sample with a similar fraction of non-hexagonal polygons to that of the metal-doped samples was prepared by electron irradiation for 20 s at an acceleration voltage of 200 kV with a dose rate of $1.5 \times 10^5 \text{ e}^- \text{ nm}^{-2} \text{ s}^{-1}$. Subsequently, the irradiated undoped sample was observed under the same conditions as the metal-doped ones. In this study, TEM observations were performed under both under-focus and over-focus conditions, and images with high visibility of dopant atoms were selected for subsequent analysis.

The PH analysis was performed with the open-source software HomCloud.³² PH visualizes the topological features of data through a persistence diagram (PD), which is a two-dimensional histogram plotting the birth (b) and death (d) scales of the topological features (holes). Each point on the PD is referred to as a birth-death (b - d) pair, and the difference $|d-b|$ is defined as the lifetime. We used a unified approach to analyze both the local and global structures of metal-doped graphene (Fig. 1). In our previous work using the developed PH-based strategy, we determined the polygon number N , representing the size of the carbon atom polygons, and quantified the connectivity between these units by counting the number of N -polygons within defect clusters.²⁹ In this study, we introduced new procedures in Figs. 1(b) and 1(c) to enable semi-automated detection of dopant positions. Under the under-focus condition shown in the filtered input image [Fig. 1(a)], metal atoms appear as dark spots surrounded by bright regions. From a topological perspective, dopant positions ideally correspond to the darkest points inside bright loops, which can be extracted as the death of a loop via filtration (here, superlevel set filtration). Therefore, we successfully automated metal atom identification [Fig. 1(c)] by performing inverse analysis of the significant b - d pairs exhibiting a lifetime greater than 40 in the one-dimensional grayscale PD [g-PD, Fig. 1(b)]. Here, superlevel and sublevel set filtrations were applied for the under-focus and over-focus images, respectively. We confirmed that the analysis results were consistent under both imaging conditions.

HR-TEM images of the four distinct graphene samples are shown in Figs. 2(a)–2(d). Because the acceleration voltage of 80 kV used during the observation was below the knock-on threshold for carbon atoms, the as-prepared pristine graphene exhibited a defect-free structure consisting solely of hexagons [Fig. 2(a)]. Furthermore, the structure showed negligible changes without defect generation even after continuous electron irradiation at 80 kV for 30 s [Fig. 2(e)]. In contrast, when graphene was doped with metal atoms—Cu in Fig. 2(b) and Cr in Fig. 2(c)—the surrounding lattice failed to maintain only hexagonal structures, resulting in the formation of non-hexagonal polygons (i.e., pentagons to octagons). Furthermore, defect reconstruction was observed even under 80 kV irradiation despite being below the knock-on threshold of graphene [Figs. 2(f) and 2(g)]. Although it is challenging to determine whether these defects originated during the deposition process or were induced by subsequent electron irradiation, it is noteworthy that the deposition and observation conditions were identical for Cu and Cr. In addition, a comparative observation of regions with few dopants was performed within the same metal-doped graphene sample under identical observation conditions, including the vacuum level and accelerating voltage

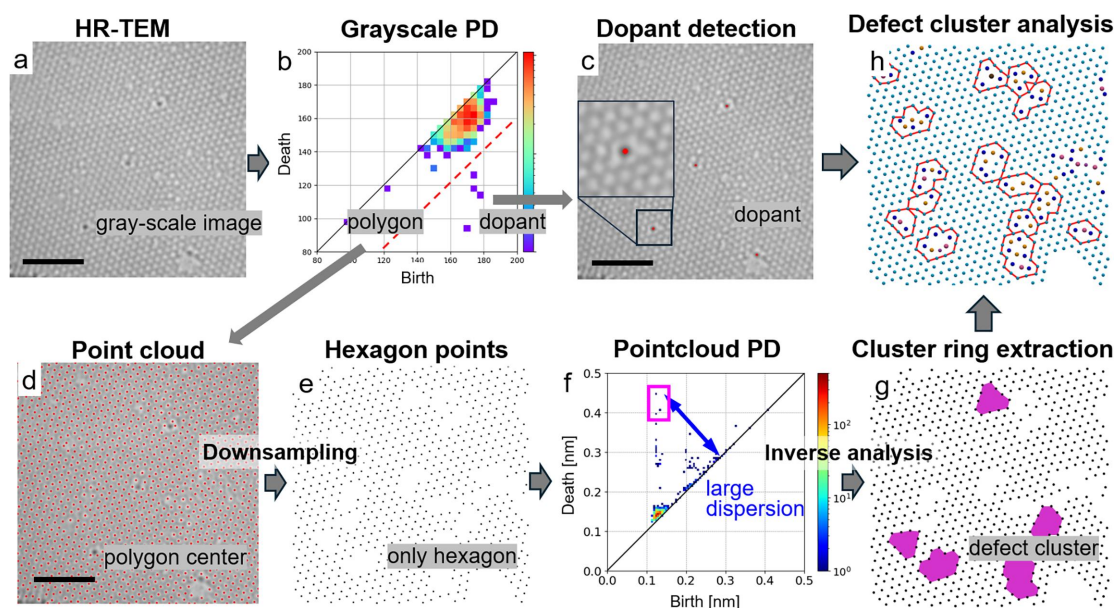


FIG. 1. Overview of the persistent homology (PH) strategy for dopant detection and defect cluster analysis. (a) Gaussian-filtered high-resolution transmission electron microscopy (HR-TEM) image ([supplementary material S1](#)) as the input for the strategy. (b) Grayscale persistence diagram (g-PD) obtained by superlevel filtration of the filtered image in (a). (c) Overlay of the extracted metal atom positions on the TEM image. (d) Overlay of the point cloud with the highest intensity within the vacuum region surrounded by carbon atom polygons. (e) Downsampled point cloud without polygon number ($N=6$). (f) One-dimensional point-cloud persistence diagram (p-PD) of the downsampled point cloud in (e). (g) Rings obtained by inverse analysis of the one-dimensional p-PD in (f). (h) Defect cluster detection using the rings in (g). The red rings indicate defect clusters. Scalebars indicate 2 nm.

([supplementary materials S2 and S3](#)). The results showed that defect generation was negligible in the regions containing few Cu and Cr atoms, suggesting that the primary cause of the observed defect generation was not residual adsorbates from sample preparation but rather the dopants. These changes are not attributed to direct knock-on damage to the pristine lattice, but to the introduction of metal atoms reducing the carbon-carbon (C-C) bond energy in the vicinity of the metal-carbon (M-C) bonds.³⁰ This energy reduction lowers the activation barrier for structural modification, thereby facilitating the displacement of C atoms or the rotation of C-C bonds even under low-voltage irradiation, leading to defect generation.

However, the rate of defect dynamics differed significantly between the Cu- and Cr-doped graphene samples. The structure around the Cr atoms rapidly evolved (on the order of tens of seconds), whereas the vicinity of the Cu atoms remained unchanged even after 150 s and subsequently evolved much more slowly (on the order of tens of minutes), as shown in [supplementary material S4](#). The difference in the rate of the structural dynamics can be attributed to three factors: (i) the displacement threshold of carbon atoms adjacent to the metal dopant, (ii) the kinetic energy transferred to the metal atom via elastic electron scattering (knock-on effect), and (iii) the difference in M-C bond strength. Generally, doping of metal atoms into the graphene lattice reduces the displacement threshold of the adjacent carbon atoms, which facilitates the structural displacement of the surrounding carbon atoms. In addition, in graphene, where knock-on damage via elastic scattering is dominant, the maximum energy transfer from an electron to a nucleus is inversely proportional to the atomic mass.¹⁵ Consequently, because Cr is lighter than Cu, it receives a larger amount of energy from the electron

beam, potentially facilitating more pronounced displacement of the surrounding atoms. Furthermore, density functional theory calculations have indicated that the Cr-C bond is stronger than the Cu-C bond.³⁰ This suggests that Cr is not only more likely to overcome the energy barrier for structural change but also tends to “drag” the surrounding C atoms during its movement owing to the stronger Cr-C bonding. Therefore, it is inferred that defect reconstruction proceeds more rapidly around Cr atoms than around Cu atoms.

To investigate the local defect structures in detail, we analyzed time-resolved snapshot HR-TEM images capturing the structural dynamics. Following the strategy outlined in [Fig. 1](#), we computed the PDs, constructed Voronoi diagrams, and calculated the polygon number for each atomic configuration. We first identified the dopant atoms; however, they exhibited negligible displacement within the observation range. The structural reconstruction around the Cu atoms was extremely slow. Therefore, we mainly compared Cr-doped graphene and undoped graphene irradiated with a 200 kV electron beam, both exhibiting rapid rates of defect dynamics [[Figs. 2\(g\) and 2\(h\)](#)]. Atomic model diagrams colored based on the calculated polygon numbers are shown in [Figs. 3\(a\)–3\(f\)](#). The corresponding original TEM images are presented in [supplementary material S5](#). For Cr-doped graphene, [Figs. 3\(a\)–3\(c\)](#) show the structures of Cr-doped graphene captured at 5, 115, and 337 s after the start of TEM observation, respectively. The three Cr atoms present in the area are indicated by red dots. Within the hexagonal graphene lattice (green), the local geometries around the NN and NNN sites of the Cr atoms were distorted, resulting in the formation of pentagons, heptagons, and octagons. [Figures 3\(d\)–3\(f\)](#) show structural models of undoped graphene, where defects were initially induced by a 200 kV acceleration voltage.

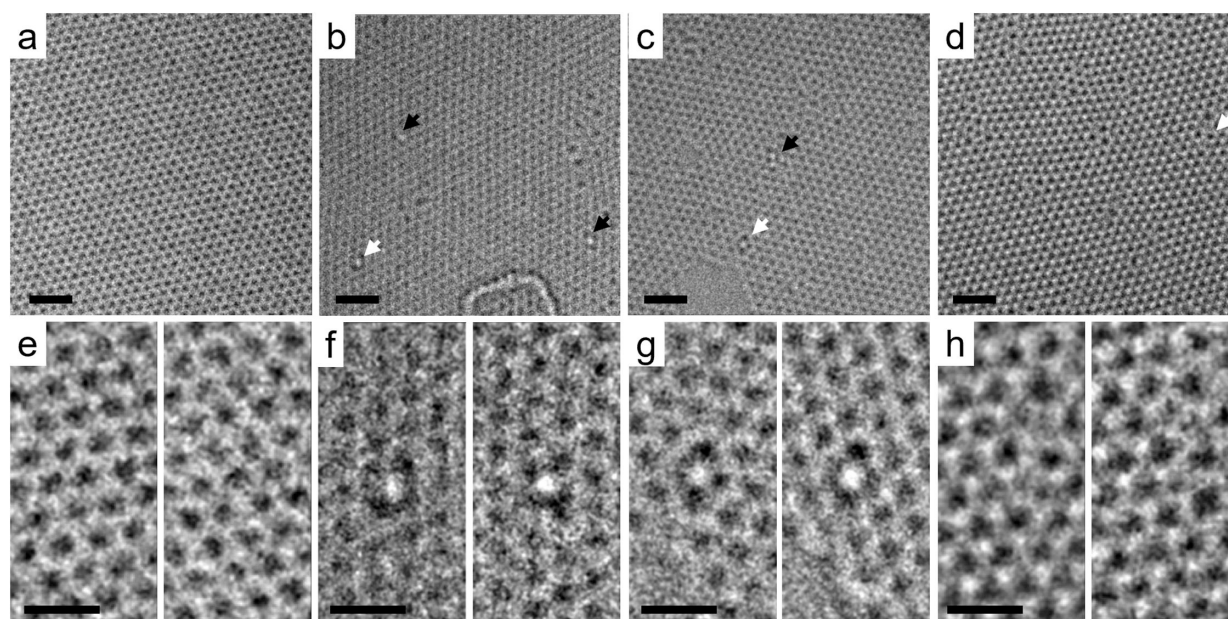


FIG. 2. HR-TEM images of various defects in graphene with/without metal atoms taken at 80 kV. (a) As-prepared defect-free graphene. (b) Cu-doped graphene (the Cu atoms are indicated by arrows) with defects. (c) Cr-doped graphene (the Cr atoms are indicated by arrows) with defects. (d) Defective graphene without metal atoms after 200 kV electron irradiation for 20 s. (e) Magnified images of as-prepared graphene from (a) (left) and as-prepared graphene after 80 kV electron irradiation for 120 s (right). (f) Magnified images of graphene around a Cu atom indicated by the white arrow in (b) (left) and Cu-doped graphene after 80 kV electron irradiation for 100 s (right). (g) Magnified images of graphene around the Cr atom indicated by the white arrow in (c) (left) and Cr-doped graphene after 80 kV electron irradiation for 20 s (right). (h) Magnified images of defective graphene indicated by the white arrow in (d) (left) and defective graphene after 80 kV electron irradiation for 50 s (right). Scale bars: (a)–(d) 1 nm and (e)–(h) 0.5 nm.

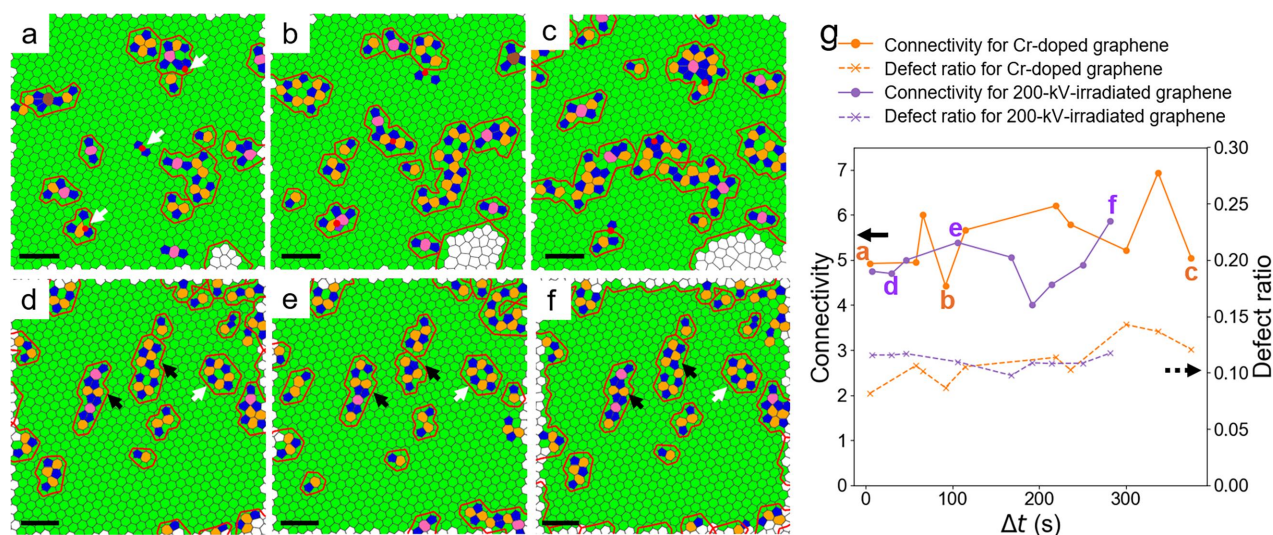


FIG. 3. Structural changes of the defect clusters in graphene introduced by Cr atoms and 200 kV electron irradiation. (a)–(f) Atomic models of graphene with defect clusters assigned from the HR-TEM images by PH. Blue, green, yellow, and pink regions correspond to pentagons, hexagons, heptagons, and octagons, respectively. Red rings indicate defect clusters. (a)–(c) Structural dynamics of Cr-doped graphene under electron beam irradiation. The red dots indicated by the arrows represent the positions of Cr atoms. The snapshots show the structures (a) 5, (b) 115, and (c) 337 s after the start of TEM image acquisition. (d)–(f) Structural dynamics of graphene with defects introduced by electron beam irradiation at 200 kV. The snapshots show the structures (d) 30, (e) 106, and (f) 282 s after the start of image acquisition. (g) Temporal changes of the connectivity and defect ratio under electron beam irradiation. Δt denotes the elapsed time since the start of image acquisition. $\Delta t = 0$ corresponds to the start of image acquisition. The data for “Cr-doped graphene” and “200 kV-irradiated graphene” represent the temporal changes in the same fields of view as those shown in (a)–(c) and (d)–(f), respectively. Scalebars indicate 1 nm.

These images correspond to 30, 168, and 282 s after the start of image acquisition. In both Cr-doped and undoped graphene with 200 kV irradiation, we observed changes not only in the numerical distribution but also in the spatial distribution of the N -polygons. Then, we calculated global defect cluster rings [red rings in Fig. 1(g)] using PH-based analysis to visualize and quantify these spatial distribution changes. Rather than tracking the individual polygon number N , the defect cluster rings in Figs. 3(a)–3(c) revealed that the clusters expanded or coalesced under electron irradiation. Furthermore, the cluster rings can accurately capture subtle structural changes. The structural dynamics of the defects in undoped graphene were less pronounced than those in Cr-doped graphene; for instance, many defect clusters composed of pentagons and heptagons, indicated by the white arrows, remained stable. In addition, the defects with pentagons and octagons indicated by the black arrows exhibited reversible changes, where the structure deformed or split [as seen in Fig. 3(e)] before reverting to its original configuration. Notably, the structural changes in defect clusters proceeded rapidly not only in the immediate vicinity of Cr atoms but also in relatively distant regions. While the aforementioned comparative experiments (supplementary materials S2 and S3) indicated that the metal dopants, rather than residual adsorbates, are the primary cause of defect generation, the influence of residual adsorbates on defect dynamics cannot be entirely excluded. Leuthner *et al.* reported that under electron irradiation, residual species such as H_2O and O_2 can transform into oxygen radicals and etch graphene, particularly in the presence of preexisting defects.³³ In contrast, no extensive etching was observed in defective regions without metal dopants in our observations, suggesting that the contribution of such adsorbates to the observed defect dynamics is limited. In addition, another plausible mechanism for the long-range defect dynamics is the relaxation of out-of-plane buckling, which can be induced by dopants or preexisting defects, even in undoped graphene. Previous studies have shown that defect clusters separated by several nanometers can undergo migration and annihilation to relieve out-of-plane buckling.³⁴ However, in our experiments, Cr-doped and undoped graphene exhibited different rates of structural reconstruction even under identical electron dose rates [see Figs. 3(a)–3(c) and 3(d)–3(f)]. Therefore, buckling relaxation is unlikely to be the primary driver of the widespread structural changes. Although our observations alone do not allow a definitive conclusion, they are consistent with the possibility that Cr doping could be a contributing factor to the long-range defect dynamics.

Next, we quantitatively evaluated the connectivity of the individual defect structures in the clusters to focus on the structural dynamics in both Cr-doped and undoped graphene with 200 kV irradiation. Specifically, we calculated the numbers of the non-hexagonal polygons within each defect cluster. The temporal changes in two metrics, the connectivity (defined as the average number of non-hexagonal polygons within a cluster) and the defect ratio (the ratio of the number of non-hexagonal polygons to the total number of polygons in the field of view), under 80 kV observation, i.e., electron beam irradiation, are summarized in Fig. 3(g). Δt denotes the elapsed time since the start of TEM image acquisition. Although the connectivity might be underestimated owing to some defect clusters being cut off at the image boundaries, the total number of polygons was fixed to 820 ± 10 for both the Cr-doped and 200 kV-irradiated graphene to ensure a valid comparison. To mitigate the boundary effects, we applied a

padding process by placing $N=6$ point groups around the perimeter.²⁹ While this process allowed us to detect clusters containing hexagons, such clusters were excluded to strictly calculate the connectivity of $N \neq 6$ polygons.

The defect ratio for Cr-doped graphene showed an increasing trend with time [yellow dashed line in Fig. 3(g)]. This is attributed to the previously mentioned increase in the number of non-hexagonal polygon with increasing irradiation time. In contrast, the defect ratio of undoped graphene irradiated at 200 kV did not change significantly with time [purple dashed line in Fig. 3(g)]. Comparing the two graphene materials, for $\Delta t \leq 100$ s, the defect ratio was lower for Cr-doped graphene. The time-averaged defect ratio (the time-averaged total number of $N \neq 6$ polygons) during this interval was 0.09 (77) for Cr-doped graphene and 0.11 (95) for irradiated graphene. However, the average connectivity within the same timeframe was 5.07 for Cr-doped graphene and 4.81 for irradiated graphene, indicating that the connectivity was higher in the Cr-doped sample. In the interval $100 \leq \Delta t \leq 250$ s, the defect ratios of the two samples were comparable. The time-averaged defect ratios (the time-averaged total number of $N \neq 6$ polygons) were 0.11 (88) for both systems. However, the time-averaged connectivity during this period was 5.46 for Cr-doped graphene and 4.76 for irradiated graphene. This indicates that local defects in Cr-doped graphene possess a strong tendency to interconnect. Furthermore, compared with the defects generated solely by 200 kV electron beam irradiation, these results suggest that the presence of Cr atoms could facilitate the formation of more spatially extended, global defect clusters. Additionally, the temporal fluctuation of the connectivity was more pronounced in Cr-doped graphene, although fluctuations were present in both cases [Fig. 3(g)]. The variance of the connectivity values in the range $0 \leq \Delta t \leq 337$ s was 0.55 for Cr-doped graphene and 0.28 for irradiated graphene. The larger fluctuation for Cr-doped graphene is consistent with the large structural changes of the defect clusters [Figs. 3(a)–3(c)]. These trends were consistently observed across other fields of view (see supplementary material S6), where the analyses were similarly standardized by the total polygon number. By incorporating defect clustering into the analysis, we successfully achieved a quantitative evaluation of the defect structure dynamics.

In summary, we have investigated the structural dynamics of defects in metal-doped graphene by combining *in situ* HR-TEM with PH-based analysis. We found that Cr doping induces more rapid structural reconstruction than Cu doping. Crucially, our PH-based analysis revealed that the defects in Cr-doped graphene possess a strong tendency to coalesce, forming highly interconnected global clusters. Quantitative comparison showed that Cr-doped graphene maintains higher defect connectivity and exhibits larger structural fluctuations than undoped graphene even at similar defect ratios. These results demonstrate that investigation of the global structure representing the connectivity of carbon polygons, in addition to the individual polygons, is effective for capturing the complex dynamics of disordered systems.

See the supplementary material for additional information: S1. Comparison between original and Gaussian-filtered HR-TEM images. S2. Regions with few Cu and Cr atoms in doped graphene. S3. Suppressed defect dynamics in a region with few Cr atoms in doped graphene. S4. Filtered HR-TEM image of Cu-doped graphene and its

time sequence. S5. Filtered HR-TEM image sequence of Cr-doped graphene and 200 kV electron-irradiated graphene. S6. Structural dynamics and connectivity analysis of defect clusters in additional regions (Region 2 and Region 3).

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

Ryuto Eguchi: Conceptualization (equal); Data curation (lead); Formal analysis (lead); Investigation (equal); Methodology (lead); Software (lead); Validation (equal); Visualization (lead); Writing – original draft (lead). **Ayako Hashimoto:** Conceptualization (equal); Funding acquisition (lead); Project administration (lead); Resources (lead); Supervision (lead); Validation (equal); Writing – review & editing (lead).

DATA AVAILABILITY

The data that support the findings of this study are available within the article and its [supplementary material](#).

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