doi:10.1017/S1431927622007097
Microsc. Microanal. 28 (Suppl 1), 2022

Probing Graphene Defect Kinetics at Millisecond Time Resolution Using Direct Detection and Machine Learning

Chen Huang[1,2], Christopher Allen[2,3], Stephen Skowron[4], Ivan Lobato[5], Takeo Sasaki[6], Sandra Van Aert[5], Elena Besley[4] and Angus Kirkland[1,2,3]

[1.] Rosalind Franklin Institute, Didcot, OX11 0FA, UK
[2.] Department of Materials, University of Oxford, Oxford, OX1 3PH, UK
[3.] Electron Physical Science Imaging Centre, Diamond Light Source, Didcot, OX11 0DE, UK
[4.] School of Chemistry, University of Nottingham, Nottingham, NG7 2RD, UK
[5.] Electron Microscopy for Materials Research (EMAT), University of Antwerp, Groenenborgerlaan 171, 2020 Antwerp, Belgium
[6.] JEOL Ltd., 3-1-2 Musashino, Akishima, Tokyo, 196-8558, Japan

Direct observation of graphene defect transitions is of great importance in studies of the chemical kinetics of topological defects in two-dimensional materials [1]. Although individual defects in graphene can now be routinely observed using transmission electron microscopy, the time resolution of these observations are typically around one second for atomic resolution TEM [2] and slower for STEM [3]. However, structural changes in graphene happen over much shorter time periods and these cannot be followed under the typical time regimes for HR(S)TEM. Slow data acquisition and traditional defect analysis methods are also restrictive in terms of the amount of defect transition processes that can be surveyed, whereas kinetic studies of defect reactions require large numbers of observations of individual reactions in order to obtain meaningful statistics.

In this work, we demonstrate direct observation of graphene defect reactions at 1ms time resolution (1000 fps) using a direct electron detector. Since the amount of data grows proportionally with increases in acquisition speed, manual analysis is practically unfeasible for the large datasets generated. Automated data analysis workflow based on a deep neural network (DNN) has been developed to identify specific defect transition processes. Using data mining of 1.8 million frames of fast HRTEM images collected in a single experimental session, more than 90000 divacancy defects (V_2(585), V_2(555-777), or V_2(5555-6-7777)) were recognised and classified using a DNN-based data processing routine.

The neural network was trained on image simulations and its validity and generality are ensured by using realistic graphene atomic models containing randomly generated defects that have been relaxed using molecular dynamics simulation as well as by the inclusion of various sources of noise and other imaging parameters corresponding to the experimental conditions. Individual frames in fast HRTEM time series are often limited by electron dose and hence have very low signal-to-noise ratio (SNR). However, the machine learning approach has proven to be capable of handling the image analysis even under low-dose conditions.

The potential applications of this method are not restricted by the specific types of defects described here and can therefore be extended to a much wider range of chemical kinetics studies in two-dimensional materials.

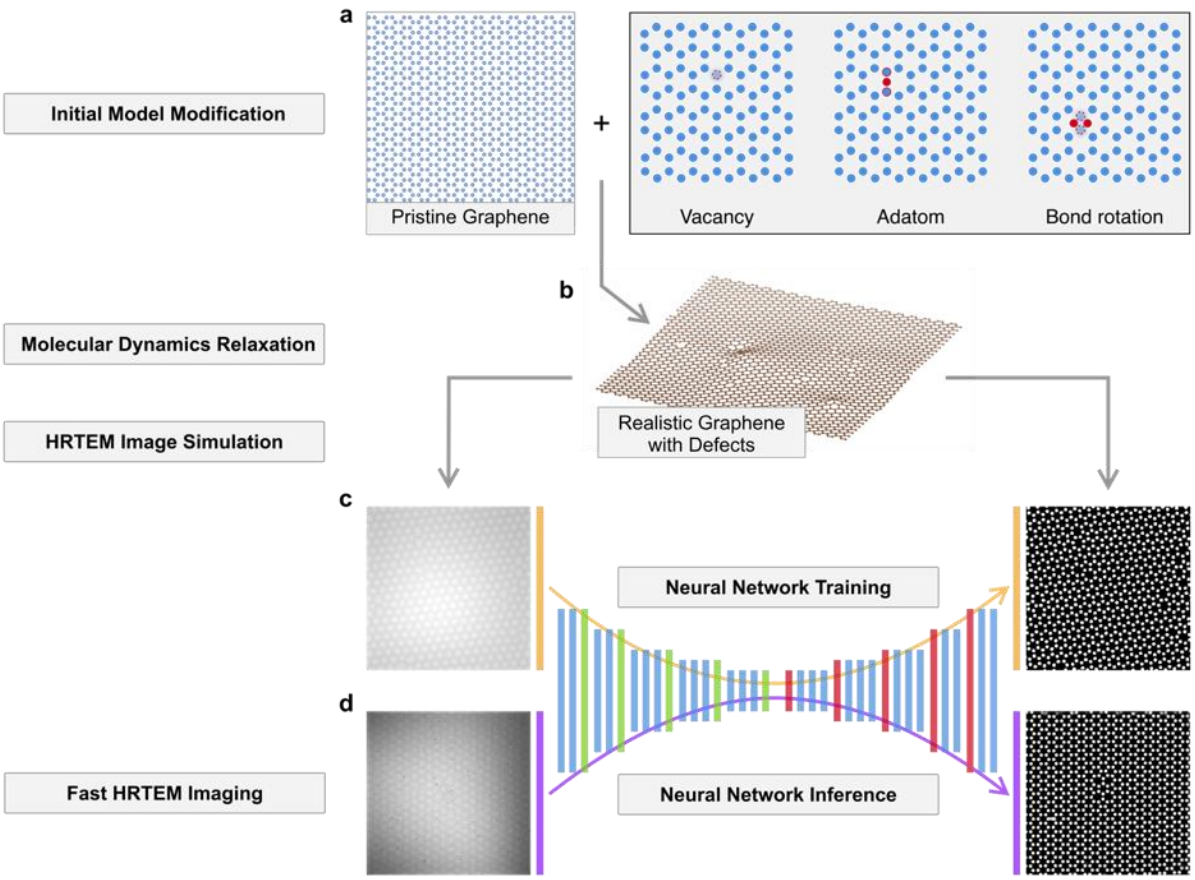

Figure 1. Illustration of the data processing strategy of DNN-based graphene defect analysis.

References

[1] ST Skowron, et al., Carbon **105** (2016), p. 176.
[2] AW Robertson et al., Nature Communications **3**(1) (2012), p. 1.
[3] J Kotakoski, C Mangler and JC Meyer, Nature Communications **5** (2014), p. 3991.

https://doi.org/10.1017/S1431927622007097 Published online by Cambridge University Press

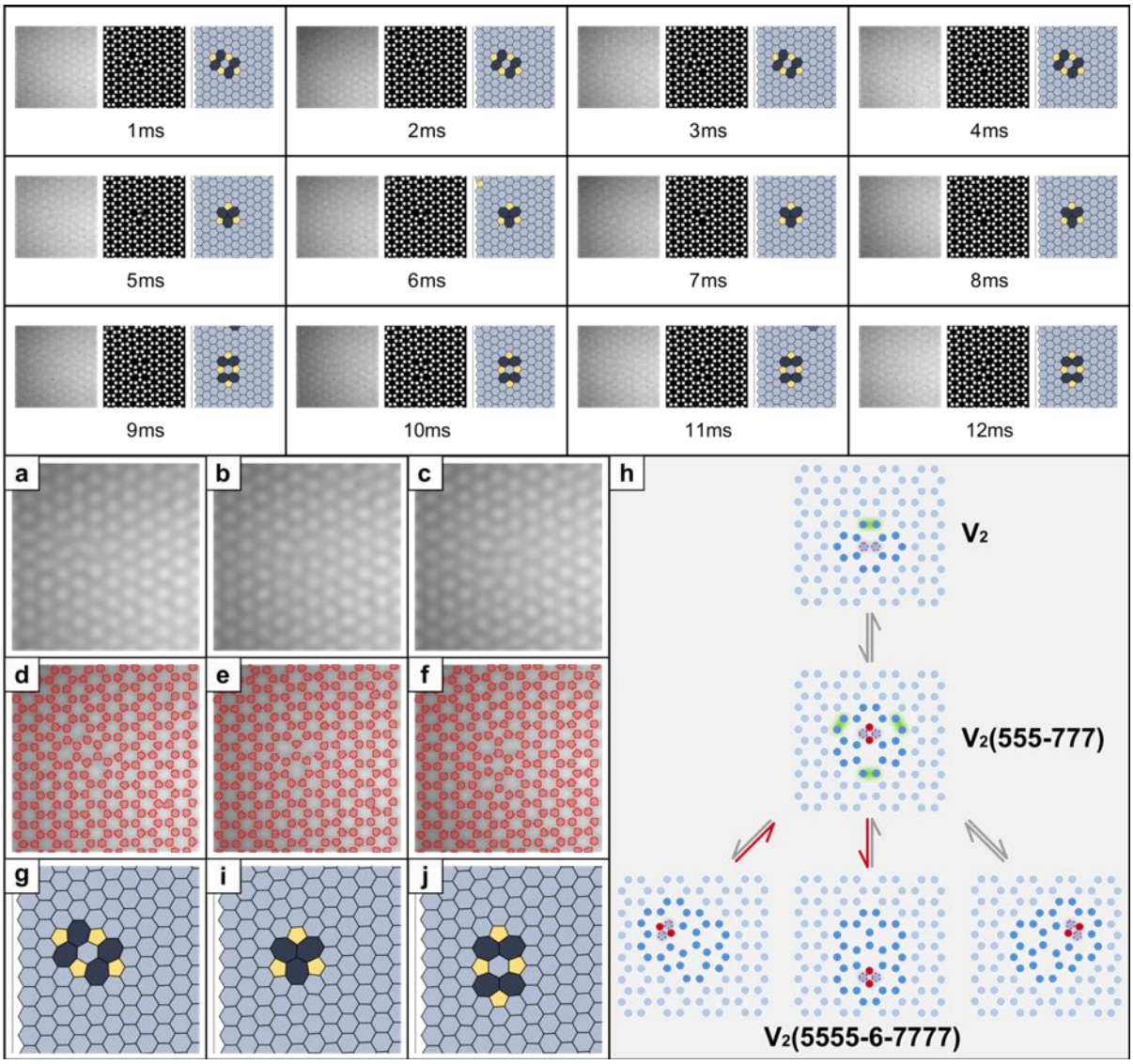

Figure 2. An example of graphene defect transition analysis over a continuous period of 12ms, showing the rotation of a V_2(5555-6-7777) via a V_2(555-777) intermediate state.

https://doi.org/10.1017/S1431927622007097 Published online by Cambridge University Press