

MATERIALS SCIENCE

Exploring order parameters and dynamic processes in disordered systems via variational autoencoders

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We suggest and implement an approach for the bottom-up description of systems undergoing large-scale structural changes and chemical transformations from dynamic atomically resolved imaging data, where only partial or uncertain data on atomic positions are available. This approach is predicated on the synergy of two concepts, the parsimony of physical descriptors and general rotational invariance of noncrystalline solids, and is implemented using a rotationally invariant extension of the variational autoencoder applied to semantically segmented atom-resolved data seeking the most effective reduced representation for the system that still contains the maximum amount of original information. This approach allowed us to explore the dynamic evolution of electron beam-induced processes in a silicon-doped graphene system, but it can be also applied for a much broader range of atomic scale and mesoscopic phenomena to introduce the bottom-up order parameters and explore their dynamics with time and in response to external stimuli.

INTRODUCTION

Over the past two decades, scanning transmission electron microscopy (STEM) has emerged as an indispensable tool for exploring materials structure and functionality on the atomic level (1–4). Recent advances in aberration correction enabled determining the position of individual atomic columns in three-dimensional (3D) materials (5) and single atoms in layered materials (6, 7) with picometer-level precision (8), enabling visualization of ferroelectric polarization and octahedral tilting fields in perovskites (9–11), symmetry-breaking phenomena in complex materials such as ferroelectric relaxors and Kitaev materials (12), and strain fields in multicomponent systems or in the presence of structural defects (13). Equally impressive are the advances in the spectroscopic modes such as electron energy loss spectroscopy, where advances ranging from single-atom spectroscopy (14, 15) to mapping plasmonic excitations and phonons have been demonstrated (14, 16–21). Last, 4D-STEM methods now offer a pathway to further increasing the information limit in STEM (22–24), providing yet unseen details of atomic-level fields and functionalities (22, 25–27).

The intriguing and yet largely unexplored opportunity presented by recent progress in STEM is the exploration of atomic-scale chemical processes, either induced by classical global stimuli like temperature or induced by electron beam (e-beam) irradiation. Notably, e-beam damage in electron microscopy has been known from the earliest days of the field (28–30) and is traditionally perceived as a strongly deleterious effect. Minimizing e-beam damage along with increasing the spatial resolution were major factors driving the development of electron microscopy instrumentation. The advent of aberration correction and the potential for atomic-resolution imaging at low voltages that it has unlocked have enabled opportunities toward systematic studies of e-beam-induced reactions. Early examples of these studies include phase transformations in 3D materials (31–34) and e-beam-induced radiolysis (35). A particularly broad

set of opportunities has emerged with extensive studies of 2D materials, where nearly all atomic units are available for observation (4) and multiple studies of vacancy and defect formation (36–41) and of uncontrolled and controlled atomic motion (42–51) have been reported. Harnessing the e-beam effects on atomic dynamics opened a pathway toward atom by atom manipulation, and recently, homo- and heteroatomic cluster assemblies have been reported (52–55).

However, further progress in the field necessitates a quantitative description of e-beam-induced transformations as a necessary step for mapping e-beam-induced reaction networks, comparison with predictive and descriptive theory, and ultimately enabling control of e-beam-induced transformations. In cases where the e-beam-induced changes are localized on a single-atom level, these descriptions are possible using the classical point defect chemistry approach, as demonstrated, for example, by Kirkland and co-workers (36, 39) for graphene and Maksov *et al.* (56) for layered transition metal dichalcogenides. However, the analysis becomes considerably more complicated in cases where large-scale changes in the connectivity of the chemical bond network occur, including changes in coordination, formation of defect agglomerates, and new phase formation. This is unsurprising because the description of the structure of disordered or partially disordered systems, ranging from amorphous solids to physical systems such as spin and cluster glasses or phase-separated oxides, has remained one of the most complex areas of condensed matter physics (57–59).

In chemically ordered systems, i.e., in cases where the chemical bond network is that of an ideal solid, symmetry-breaking phenomena can be effectively analyzed using machine learning tools such as multivariate statistical methods or more complex autoencoder (AE)- or variational AE (VAE)-based approaches. In this case, symmetry-breaking distortions are referenced to the known (from the global average) ideal lattice, allowing for straightforward analysis. Recently, this approach was applied to the structural description of perovskites based on column (11, 60) and unit cell (61) shape analysis, as well as ferroelectric materials and layered chalcogenides based on the analysis of local atomic neighborhoods (62). However, this approach fails when the chemical bonding network is nonperiodic, precluding the unambiguous definition of the high-symmetry state.

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