

## MATERIALS SCIENCE

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

Sergei V. Kalinin<sup>1\*</sup>, Ondrej Dyck<sup>1</sup>, Stephen Jesse<sup>1</sup>, Maxim Ziatdinov<sup>2\*</sup>

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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# Dynamical Processes In Disordered Systems

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## **Dynamical Processes In Disordered Systems:**

**Dynamical Processes in Disordered Systems** William M. Yen, 1990-01-01 Materials Science Forum Vol 51

### **Dynamical Processes In Condensed Molecular Systems - Proceedings Of The Emil-warburg Symposium A**

Blumen, Joseph Klafter, D Haarer, 1990-12-19 The research on condensed molecular solids is truly interdisciplinary spanning the range from statistical and molecular physics to solid state physics chemistry up to materials science This Symposium on dynamical processes in condensed molecular systems highlights the most recent developments in the field focussing on low dimensional and non crystalline materials such as Langmuir Blodgett films polymers and glasses The text includes both advanced experimental techniques hole burning fluorescence short time pulses nonlinear spectroscopy and also modern theoretical approaches dynamical percolation fractals localization

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**Disordered Systems**, 1994

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Blumen, Joseph Klafter, Joshua Jortner, 1989-05-01 This review volume provides an up to date review of experimental methods and theoretical approaches in the study of dynamical processes in condensed molecular systems The experimental contributions include hole burning in glasses and in proteins optical dephasing in glasses photo conductivity in polymers energy transfer among molecules in confining spaces and electron transfer in polar solvents The theoretical part summarizes recent advances on hole burning hierarchical aspects of relaxation and transport in disordered systems

### **Dynamical Processes on Complex Networks**

Alain Barrat, Marc Barthélemy, Alessandro Vespignani, 2008-10-23 The availability of large data sets has allowed researchers to uncover complex properties such as large scale fluctuations and heterogeneities in many networks leading to the breakdown of standard theoretical frameworks and models Until recently these systems were considered as haphazard sets of points and connections Recent advances have generated a vigorous research effort in understanding the effect of complex connectivity patterns on dynamical phenomena This book presents a comprehensive account of these effects A vast number of systems from the brain to ecosystems power grids and the internet can be represented as large complex networks This book will interest graduate students and researchers in many disciplines from physics and statistical mechanics to mathematical biology and information science Its modular approach allows readers to readily access the sections of most interest to them and complicated maths is avoided so the text can be easily followed by non experts in the subject

**Dynamical Processes in Molecular Physics, Lectures from the first EPS Southern European School of Physics, Avila, September 1991** Gerardo Delgado-Barrio, 1993-01-01 In Dynamical Processes in Molecular Physics leading European lecturers outline the fundamental aspects of dynamical processes in molecular physics The papers included in this book make a valuable contribution to the teaching of molecular physics as well as discussing advances in this area It covers a wide range of interesting and relevant topics both experimental and theoretical Physicists and physical chemists at graduate and research level will find this an invaluable reference

**Disordered Solids** Baldassare Di Bartolo, 2013-03-09 This book

presents an account of the course Disordered Solids Structures and Processes held in Erice Italy from June 15 to 29 1987 This meeting was organized by the International School of Atomic and Molecular Spectroscopy of the Ettore Majorana Centre for Scientific Culture The objective of this course was to present the advances in physical modelling mathematical formalism and experimental techniques relevant to the interpretation of the structures of disordered solids and of the physical processes occurring therein Traditional solid state physics treats solids as perfect crystals and takes great advantage of their symmetry by means of such mathematical formalisms as the reciprocal lattice the Brillouin zone and the powerful tools of group theory Even if in reality no solid is a perfect crystal this theoretical approach has been of great usefulness in describing solids deviations from perfect order have been treated as perturbations of the ideal model A new situation arises with truly disordered solids where any vestige of long range order has disappeared The basic problem is that of describing these systems and gaining a scientific understanding of their physical properties without the mathematical formalism of traditional solid state physics While some of the old approaches may occasionally remain valid e g chemical bonding approach for amorphous solids the old ways will not do Disorder is not a perturbation with disorder something basically new may be expected to appear

#### **Disorder Effects on Relaxational Processes** Ranko Richert,Alexander Blumen,2012-12-06

The field of non crystalline materials has seen the emergence of many challenging problems during its long history In recent years the interest in polymeric and biological disordered matter has stimulated new activities which in turn have enlarged the organic and inorganic glass community The current research fields and recent progress have extended our knowledge of the rich phenomenology of glassy systems where the role of disorder is fundamental for the underlying microscopic dynamics In addition despite the lack of a unified theory many interesting theoretical models have recently evolved The present volume offers the reader a collection of topics representing the current state in the understanding of disorder effects as well as a survey of the basic problems and phenomena involved The task of compiling a book devoted to disordered systems has benefited much from a seminar organized by the W E Heraeus Foundation in Bad Honnef in April 1992 where we had the opportunity to discuss the project with most of the authors Here we wish to thank the Heraeus Foundation for their support and the authors and Springer Verlag especially Dr Marion Hertel for the pleasant cooperation

*Scaling And Disordered Systems: International Workshop And Collection Of Articles Honoring Professor Antonio Coniglio On The Occasion Of His 60th Birthday* Mohamed Daoud,Fereydoon Family,Hans J Herrmann,H Eugene Stanley,2002-05-30

Investigation of the fractal and scaling properties of disordered systems has recently become a focus of great interest in research Disordered or amorphous materials like glasses polymers gels colloids ceramic superconductors and random alloys or magnets do not have a homogeneous microscopic structure The microscopic environment varies randomly from site to site in the system and this randomness adds to the complexity and the richness of the properties of these materials A particularly challenging aspect of random systems is their dynamical behavior Relaxation in disordered systems generally follows an

unusual time dependent trajectory Applications of scaling and fractal concepts in disordered systems have become a broad area of interdisciplinary research involving studies of the physics chemistry mathematics biology and engineering aspects of random systems This book is intended for specialists as well as graduate and postdoctoral students working in condensed matter or statistical physics It provides state of the art information on the latest developments in this important and timely topic The book is divided into three parts Part I deals with critical phenomena Part II is devoted to discussion of slow dynamics and Part III involves the application of scaling concepts to random systems The effects of disorder at the mesoscopic scale as well as the latest results on the dynamical properties of disordered systems are presented In particular recent developments in static and dynamic scaling theories and applications of fractal concepts to disordered systems are discussed

**Optical Studies of Dynamical Processes in Disordered Systems. Progress Report** ,1994 The authors present an abbreviated summary of the progress they have attained in the course of the abbreviated first year of the present three year grant The focus of their research continues to be on studies which help them understand various dynamical processes which affect the structure and the optical properties of disordered and amorphous materials They continue to make significant progress in their attempts to understand the factors which affect for example the efficiencies of activated glasses This report contains a brief description of the work they have carried out during the present grant period and an outline of the initiatives they are presently undertaking or continuing during the second period Proceedings of the ... International Conference on Dynamical Processes in the Excited States of Solids ,1985

**Energy Research Abstracts** ,1977 Semiannual with semiannual and annual indexes References to all scientific and technical literature coming from DOE its laboratories energy centers and contractors Includes all works deriving from DOE other related government sponsored information and foreign nonnuclear information Arranged under 39 categories e g Biomedical sciences basic studies Biomedical sciences applied studies Health and safety and Fusion energy Entry gives bibliographical information and abstract Corporate author subject report number indexes *Physics Briefs* ,1994

**Optical Studies of Dynamical Processes in Disordered Systems. [Annual] Progress Report** ,2005 Focus continues to be on dynamical processes such as relaxation and energy diffusion which affect the structure and optical properties of disordered and amorphous materials particularly glasses which are luminescent in the near infrared and their efficiency Elementary excitations in amorphous materials are also under study 12 refs *Advanced ESR Methods in Polymer Research* Shulamith Schlick,2006-09-18 This one of a kind book introduces the fundamentals of ESR to polymer scientists while focusing on the significance of recent advanced ESR methods for polymeric systems The Fundamentals section provides information on ESR spectra experimental techniques and data analysis

**Optical Studies of Dynamical Processes in Disordered Systems. [Annual] Progress Report** ,1993 Focus continues to be on dynamical processes such as relaxation and energy diffusion which affect the structure and optical properties of disordered and amorphous materials particularly glasses which are luminescent in the

near infrared and their efficiency Elementary excitations in amorphous materials are also under study 12 refs

*International Conference on Disorder and Chaos* A. Vulpiani, 1998      **Faculties, Publications, and Doctoral Theses in Chemistry and Chemical Engineering at United States Universities** American Chemical Society. Committee on Professional Training, 1991      Dissertationes Mathematicae , 2005      Land Evaluation George Alan Stewart, 1968 Papeles del symposium del csiro celebrado en agosto de 1968 en los que se estudian los principios de clasificacion y evaluacion del terreno en funcion de sus recursos naturales demanda de los usuarios productividad biologica etc Estableciendose los criterios para elaboracion e interpretacion de datos asi como los parametros de evaluacion

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