

# Prediction of Electronic Properties of Radical-Containing Polymers at Coarse-Grained Resolutions

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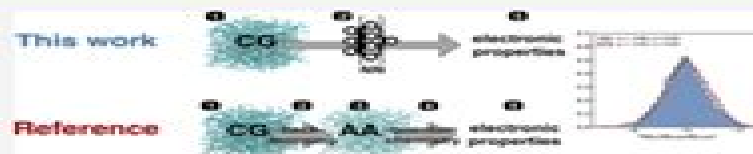


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**ABSTRACT:** The properties of soft electronic materials depend on the coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales. The description of such properties requires multiscale approaches capable of, at the same time, accessing electronic properties and sampling the conformational space of soft materials. This could in principle be realized by connecting the coarse-grained (CG) methodologies required for adequate conformational sampling to conformationally averaged electronic property distributions via backmapping to atomistic-resolution level models and repeated quantum-chemical calculations. Computational demands of such approaches, however, have hindered their application in high-throughput computer-aided soft materials discovery. Here, we present a method that, combining machine learning and CG techniques, can replace traditional backmapping-based approaches without sacrificing accuracy. We illustrate the method for an emerging class of soft electronic materials, namely, nonconjugated, radical-containing polymers, promising materials for all-organic energy storage. Supervised machine learning models are trained to learn the dependence of electronic properties on polymer conformation at CG resolutions. We then parametrize CG models that retain electronic structure information, simulate CG condensed phases, and predict the electronic properties of such phases solely from the CG degrees of freedom. We validate our method by comparing it against a full backmapping-based approach and find good agreement between both methods. This work demonstrates the potential of the proposed method to accelerate multiscale workflows and provides a framework for the development of CG models that retain electronic structure information.



## INTRODUCTION

Radical-containing polymers, also known as open-shell macromolecules, macromolecular radicals, or simply radical polymers, possess intriguing redox, optoelectronic, and magnetic characteristics that make them appealing for applications ranging from energy storage and optoelectronics to spintronics and memory storage.<sup>1–6</sup> Nonconjugated, radical-containing polymers are organic polymers that have a nonconjugated backbone bearing pendant stable radical sites. They constitute a class of charge-carrying polymers that do not rely on  $\pi$ -conjugation to transport charges successfully. The rational design of radical polymers with enhanced characteristics could be greatly advanced by deriving relationships that connect their molecular structure, morphology, and electronic properties. These relationships are inherently multiscale, involving the coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales. New modeling approaches capable of describing such coupling are needed.

Recent work has shown that coarse-grained (CG) models can be used to probe polymeric material length and time scales reaching the mesoscale.<sup>7–10</sup> In contrast, explicit quantum-chemical calculations, usually using density functional theory (DFT), which are necessary to access electronic properties,<sup>11–13</sup> are extraordinarily demanding and can only capture picosecond and Angstrom-level processes. To bridge these two

scales, that is, to take into account large-scale morphological features generated via self-assembly processes (possibly as a function of processing conditions) when computing electronic properties, recent efforts have sought to introduce new multiscale modeling approaches.<sup>12,14</sup> In such approaches, the soft material morphology generated via CG simulations is backmapped to the atomistic resolution required for the quantum-chemical calculations, and such calculations are then performed on conformations drawn from the backmapped morphologies. These multiscale approaches have been primarily developed in the context of organic semiconductors<sup>12,15–17</sup> and provide a means to access the conformational dependence of electronic properties. However, considerable computational demands and workflow complexity of such have limited their applicability for high-throughput computer-aided materials discovery.

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# Electronic Properties Of Polymers

**H. Kuzmany, M. Mehring, Siegmund Roth**

A red circular graphic with a gradient, appearing as a semi-circle or a partial circle, located to the right of the authors' names.

## **Electronic Properties Of Polymers:**

**Electrical Properties of Polymers** A. R. Blythe, David Bloor, 2005-06-10 Fully revised and expanded this second edition of A Blythe's successful title on electrical properties of polymers covers both the fundamental and recent developments in this growing area. This book provides a broad and comprehensive account on the topic describing underlying physical principles and synthesis through to emerging technologies. The new edition provides particular emphasis to the new generation of conductive polymers. Emerging uses of polymers in industrial applications are described and cover topics such as light emitting diodes, flexible polymers and soft electronics. Written in an accessible style without complicated theory, this book combines key concepts with applications. With the inclusion of further reading material provided at the end of each chapter for interested readers, this book is an authoritative guide to advanced level undergraduates and graduates studying polymer materials and physical sciences. It will also be of significant interest to researchers working in this evolving field.

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research and applications in the field Like the previous winter schools on polymers this one concentrated on the electronic structure and potential for application of polymers with conjugated double bonds This time however special attention was paid to the effects of orientation and dimensionality Anisotropy of the electric conductivity in stretch oriented samples and whether the transport mechanisms are one two or three dimensional or might even have a fractal dimensionality were there fore central topics The problem of orientation was extended to systems such as Langmuir Blodgett films and other layered structures Accordingly thin films were the focus of most of the application oriented contributions Whereas in the previous winter schools discussions on applications dealt with large volume applications such as electromagnetic shielding and energy storage this time molecular materials for electronics and prospects of molecular electronics were at the center of interest

*Optical and Electrical Properties of Polymers: Volume 214* John A. Emerson, John M. Torkelson, 1991-10-18 The MRS Symposium Proceeding series is an internationally recognised reference suitable for researchers and practitioners

**Electronic Properties of Polymers and Related Compounds** H. Kuzmany, M. Mehring, Siegmund Roth, 2012-12-06 At the International Winter School on Electronic Properties of Polymers and Related Compounds particular attention was paid to a very new and special field in polymer research It is concerned with the study of the electronic structure of polymers and with physical and chemical properties directly related to this structure In particular tutorial and research contributions on electrical electrochemical optical magnetic lattice dynamical and structural properties were presented In addition review reports on related topics such as charge transfer complexes and linear chain compounds transition metal trichalcogenides were included In two discussion meetings the special role of polyacetylene and possible present and future applications of the electronic properties of polymers as e g conductors or as electrodes in electrochemical cells were elucidated The electronic properties of polymers cover a wide range of research problems which are of particular interest for polymers with a  $\pi$ -electron system Thus a great part of the work presented was concerned with conjugated systems Additional presentations dealt with other systems such as biopolymers photopolymers or electrets which are of significant scientific and technical importance It was demonstrated how their electronic properties are increasingly being investigated from a fundamental point of view by applying known concepts of solid state science *Electronic Properties of Polymers* J Mort (ed), 1982 [Polymers for Electricity and Electronics](#) Jiri George Drobny, 2012-02-07 The comprehensive practical book that explores the principles properties and applications of electrical polymers The electrical properties of polymers present almost limitless possibilities for industrial research and development and this book provides an in depth look at these remarkable molecules In addition to traditional applications in insulating materials wires and cables electrical polymers are increasingly being used in a range of emerging technologies Presenting a comprehensive overview of how electrical polymers function and how they can be applied in the electronics automotive medical and military fields *Polymers for Electricity and Electronics Materials Properties and Applications* presents intensive and accessible coverage with a focus on practical applications

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**Electronic Properties of Conjugated Polymers** Hans Kuzmany, Michael Mehring, Siegmund Roth, 2012-12-06 This book deals with electrical electrochemical structural magnetic optical and lattice dynamical properties of conjugated polymers such as polyaniline polyacetylene polydiacetylene polypyrrole polyparaphenylene and polythiophene Several new conjugated systems and model polyenes are also considered Since the previous winter school on this topic held in 1985 the focus of interest in the field has broadened and now covers not only conductivity and relaxation phenomena of polyacetylene but also nonlinear optical properties highly oriented and single crystal polymers and electrochemical and opto electrochemical properties of special materials Particular attention is paid in this volume to the possible applications of these systems for example in electrochemical cells as electrode materials and in nonlinear optics devices which now appear to be much more realistic than previously The detailed contributions are complemented by short reviews of thin film polymers Langmuir Blodgett layers filled polymers ferromagnetic polymers superconducting low dimensional systems including organic superconductors and high temperature superconductors and the application of fractal models to polymers

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Electronic Properties of Polymers and Related Compounds Hans Kuzmany, M. Mehring, Siegmund Roth, 1985 *Rapra Review Reports*, *Principles of Polymer Systems, Sixth Edition* Ferdinand Rodriguez, Claude Cohen, Christopher K. Ober, Lynden Archer, 2014-12-09 Maintaining a balance between depth and breadth the Sixth Edition of Principles of Polymer Systems continues to present an integrated approach to polymer science and engineering A classic text in the field the new edition offers a comprehensive exploration of polymers at a level geared toward upper level undergraduates and beginning graduate students Revisions to the sixth edition include A more detailed discussion of crystallization kinetics strain induced crystallization block copolymers liquid crystal polymers and gels New powerful radical polymerization methods Additional polymerization process flow sheets and discussion of the polymerization of polystyrene and poly vinyl chloride New discussions on the elongational viscosity of polymers and coarse grained bead spring molecular and tube models Updated information on models and experimental results of rubber elasticity Expanded sections on fracture of glassy and semicrystalline polymers New sections on fracture of elastomers diffusion in polymers and membrane formation New coverage of polymers from renewable resources New section on X ray methods and dielectric relaxation All chapters have been updated and out of date material removed The text contains more theoretical background for some of the fundamental concepts pertaining to polymer structure and behavior while also providing an up to date discussion of the latest developments in polymerization systems Example problems in the text help students through step by step solutions and nearly 300 end of chapter problems many new to this edition reinforce the concepts presented

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#### **Electronic Properties of Polymers and Applications** European Physical Society, Jednota československých matematiků a fyziků v Praze,

**Electronic Properties of Conjugated Polymers III**  
Hans Kuzmany, Michael Mehring, Siegmund Roth, 1989-11-02 This book deals with electrical electrochemical structural magnetic optical and lattice dynamical properties of conjugated polymers such as polyaniline polyacetylene polydiacetylene polypyrrole polyparaphenylene and polythiophene Several new conjugated systems and model polyenes are also considered Since the previous winter school on this topic held in 1985 the focus of interest in the field has broadened and now covers not only conductivity and relaxation phenomena of polyacetylene but also nonlinear optical properties highly oriented and single crystal polymers and electrochemical and opto electrochemical properties of special materials Particular attention is paid in this volume to the possible applications of these systems for example in electrochemical cells as electrode materials and in nonlinear optics devices which now appear to be much more realistic than previously The detailed contributions are complemented by short reviews of thin film polymers Langmuir Blodgett layers filled polymers ferromagnetic polymers superconducting low dimensional systems including organic superconductors and high temperature superconductors and the application of fractal models to polymers

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