

#### **Introduction to Graphene-Based Nanomaterials**

Graphene is one of the most intensively studied materials and has unusual electrical, mechanical, and thermal properties which provide almost unlimited potential applications. This book provides an introduction to the electrical and transport properties of graphene and other two-dimensional nanomaterials, covering *abinitio* to multiscale methods. Updated from the first edition, this book contains added chapters on other two-dimensional materials, spin-related phenomena, and an improved overview of Berry phase effects. Other topics include powerful order-N electronic structure, transport calculations, AC transport, and multiscale transport methodologies. Chapters are complemented with concrete examples and case studies, questions and exercises, detailed appendices, and computational codes. This book is a valuable resource for graduate students and researchers working in physics, or materials science or engineering, who are interested in the field of graphene-and two-dimensional nanomaterials.

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# Introduction to Graphene-Based Nanomaterials

## **From Electronic Structure to Quantum Transport**

Second Edition

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### **Preface to the Second Edition**

We are delighted to give a second birth to this book on the presentation of electronic and transport properties of graphene-related materials five years after its first edition. Over these years, research in the area of graphene and two-dimensional materials has evolved. The changes are well reflected by the name of the 2017 Solvay Workshop dedicated to this field: *From physics of graphene to graphene for physics*. Indeed today, rather than studying graphene's properties, we are using it mostly as a platform for unveiling new physics. Furthermore, with each newly discovered two-dimensional material a similar path emerges, from studying their properties to using them as a brave new playground for exciting physics. At the same time, we are witnessing a very interesting return to three-dimensional systems, this time based on the stacking of two-dimensional structures which allows for more tailor-made properties.

In this new edition we decided to expand different aspects that were either missing or too succint in the previous edition. This includes, the new family of two-dimensional materials Berry phases and quantum Hall effects, and spintronics. We have reduced a few chapters that did not age very well to make space for this more exciting new material. As with the first edition we keep being committed to offering problems and solutions through our website, besides those provided in the book.

Please do not hesitate to contact us if you have questions or comments. In this sense, when choosing whom to contact it might be useful to keep in mind the distribution of our contributions to the book, which take advantage of our complementary expertise: Jean-Christophe wrote of the parts dealing with *ab initio* and new two-dimensional materials (Chapters 1, 3, 10, Appendices A and B, and parts of Chapter 2 and Appendix C), Stephan was in charge of spintronics, disorder effects, and transport (Chapters 6, 8, Appendix D, and parts of Chapters 2, 4, 5, and 7), Luis took care of the transport, quantum Hall effects, and AC transport parts (Chapters 4, 7, and 9 and parts of Chapters 2, 5, and Appendix C).

We would like to thank again our colleagues and coworkers for their help and inspiring our work. Besides those already acknowledged in our first preface, here we would like to also thank those newcomers to our family: Matías Berdakin, David Beljonne, Felix Casanova, Aurélie Champagne, José Chesta, Samuel Dechamps, José Hugo García, Rocío González, Benoît Hackens, Hafid Khalfoun, Víctor Manuel Martínez Álvarez, Joaquín Medina, Viet-Hung Nguyen, Álvaro Núñez, Esteban Rodríguez, Pierre Seneor, Christoph Stampfer, Benoît Van Troeye, and Marc Vila.



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We thank our home institutions for supporting our research, as well as the Alexander von Humboldt Foundation (SR and LEFFT), the Abdus Salam International Centre for Theoretical Physics (LEFFT), and the Francqui-Stichting Foundation of Belgium (JCC). Finally, we are indebted to our respective wives (Sandra Rieger, Encarni Carrasco Perea, and Mireille Toth-Budai) and our children (Hector and Gabriel Roche, and Ilona, Elise, and Mathilde Charlier) for their warm enthusiasm and continuous support during all these years of time-consuming work.

We hope that you find this second edition to be a useful companion for starting in this field and perhaps even for your day-to-day research.

And we wish you an exciting journey in Flatland and beyond!



### **Preface to the First Edition**

Once deemed impossible to exist in nature, graphene, the first truly two-dimensional nanomaterial ever discovered, has rocketed to stardom since being first isolated in 2004 by Nobel Laureates Konstantin Novoselov and Andre K. Geim of the University of Manchester. Graphene is a single layer of carbon atoms arranged in a flat honeycomb lattice. Researchers in high energy physics, condensed matter physics, chemistry, biology, and engineering, together with funding agencies and companies from diverse industrial sectors, have all been captivated by graphene and related carbon-based materials such as carbon nanotubes and graphene nanoribbons, owing to their fascinating physical properties, potential applications, and market perspectives.

But what makes graphene so interesting? Basically, graphene has redefined the limits of what a material can do: it boasts record thermal conductivity and the highest current density at room temperature ever measured (a million times that of copper!); it is the strongest material known (a hundred times stronger than steel!) yet is highly mechanically flexible; it is the least permeable material known (not even helium atoms can pass through it!); the best transparent conductive film; the thinnest material known; and the list goes on.

A sheet of graphene can be quickly obtained by exfoliating graphite (the material that the tip of your pencil is made of) using sticky tape. Graphene can readily be observed and characterized using standard laboratory methods, and can be mass-produced either by chemical vapor deposition (CVD) or by epitaxy on silicon carbide substrates. Driven by these intriguing properties, graphene research is blossoming at an unprecedented pace and marks the point of convergence of many fields. However, given this rapid development, there is a scarcity of tutorial material to explain the basics of graphene while describing the state of the art in the field. Such materials are needed to consolidate the graphene research community and foster further progress.

The dearth of up-to-date textbooks on the electronic and transport properties of graphene is especially dramatic: the last major work of reference in this area – written by Riichiro Saito, Gene Dresselhaus, and Mildred Dresselhaus – was published in 1998. Seeking to answer the prayers of many colleagues who have had to struggle in a nascent field characterized by a huge body of research papers but very little introductory material, we decided to write this book. It is the fruit of our collective research experience, dating from the early days of research on graphene and related materials, up through the past decade, when each of us developed different computational tools and



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theoretical approaches to understanding the complex electronic and transport properties in realistic models of these materials.

We have written *Introduction to Graphene-Based Nanomaterials: From Electronic Structure to Quantum Transport* for everyone doing (or wishing to do) research on the electronic structure and transport properties of graphene-related systems. Assuming basic knowledge of solid-state physics, this book offers a detailed introduction to some of the most useful methods for simulating these properties. Furthermore, we have made additional resources (computational codes, a forum, etc.) available to our readers at cambridge.org/foatorres, and at the book website (*introductiontographene.org*), where additional exercises as well as corrections to the book text (which will surely appear) will be posted.

Graphene and related materials pertain to a larger family that encompasses all kinds of two-dimensional materials, from boron nitride lattices, to transition-metal dichalcogenides ( $MoS_2$ ,  $WS_2$ ), to the silicon analogue of graphene, silicene, a recently discovered zero-gap semiconductor. Researchers are beginning to explore the third dimension by shuffling two-dimensional materials and by fabricating three-dimensional heterostructures (BN/graphene,  $BN/MoS_2/graphene$ , etc.) with unprecedented properties.

Interestingly, low-energy excitations in two-dimensional graphene (and in one-dimensional metallic carbon nanotubes), known as massless Dirac fermions, also develop at the surface of topological insulators (such as Bi<sub>2</sub> Se<sub>3</sub>, Bi<sub>2</sub>Te<sub>3</sub>, etc.), which are bulk insulators. Topological insulators thus share commonalities with graphene, such as Berry's phase-driven quantum phenomena (Klein tunneling, weak antilocalization, etc.), and exhibit other features such as spin-momentum locking that offer different and ground-breaking perspectives for spintronics. Therefore, we believe that our presentation of the fundamentals of electronic and transport properties in graphene and related materials should prove useful to a growing community of scientists, as they touch on advanced concepts in condensed matter physics, materials science, and nanoscience and nanotechnology.

The book starts with an introduction to the electronic structures and basic concepts in transport in low-dimensional materials, and then proceeds to describe the specific transport phenomena unique to graphene-related materials. Transport concepts are then presented through simple disorder models, which in some cases enable comparison with analytical treatments. Additionally, the development of multiscale quantum transport methodologies (either within the Landauer–Büttiker or Kubo–Greenwood formalisms) is introduced in a straightforward way, showing the various options for tackling defects and impurities in graphene materials with more structural and chemical complexity: from combined *ab initio* with tight-binding models to transport calculations fully based on first principles. To facilitate reading, the essential technical aspects concerning the formalism of Green functions, as well as transport implementation and order-N transport schemes are described in dedicated appendices.

This book encompasses years of scientific research, research that has enabled us to establish certain foundations in the field, a work made possible by the efforts of



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We hope that you find this book to be a useful companion for starting in this field and perhaps even for your day-to-day research. We recommend that you start by reading Chapter 1 and then follow the advice in *Guide to the book* (Section 1.3).

And we wish you an exciting journey in Flatland!...

