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Modern Introduction to Surface Plasmons:

Theory, Mathematica Modeling and Applications

表面等离子激元现代导论

——理论、Mathematica建模与应用

（影印版）

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序　　言

物理学是研究物质、能量以及它们之间相互作用的科学。她不仅是化学、生命、材料、信息、能源和环境等相关学科的基础，同时还是许多新兴学科和交叉学科的前沿。在科技发展日新月异和国际竞争日趋激烈的今天，物理学不仅囿于基础科学和技术应用研究的范畴，而且在社会发展与人类进步的历史进程中发挥着越来越关键的作用。

我们欣喜地看到，改革开放三十多年来，随着中国政治、经济、教育、文化等领域各项事业的持续稳定发展，我国物理学取得了跨越式的进步，做出了很多为世界瞩目的研究成果。今日的中国物理正在经历一个历史上少有的黄金时代。

在我国物理学科快速发展的背景下，近年来物理学相关书籍也呈现百花齐放的良好态势，在知识传承、学术交流、人才培养等方面发挥着无可替代的作用。从另一方面看，尽管国内各出版社相继推出了一些质量很高的物理教材和图书，但系统总结物理学各门类知识和发展，深入浅出地介绍其与现代科学技术之间的渊源，并针对不同层次的读者提供有价值的教材和研究参考，仍是我国科学传播与出版界面临的一个极富挑战性的课题。

为有力推动我国物理学研究、加快相关学科的建设与发展，特别是展现近年来中国物理学者的研究水平和成果，北京大学出版社在国家出版基金的支持下推出了《中外物理学精品书系》，试图对以上难题进行大胆的尝试和探索。该书系编委会集结了数十位来自内地和香港顶尖高校及科研院所的知名专家学者。他们都是目前该领域十分活跃的专家，确保了整套丛书的权威性和前瞻性。

这套书系内容丰富，涵盖面广，可读性强，其中既有对我国传统物理学发展的梳理和总结，也有对正在蓬勃发展的物理学前沿的全面展示；既引进和介绍了世界物理学研究的发展动态，也面向国际主流领域传播中国物理的优秀专著。可以说，《中外物理学精品书系》力图完整呈现近现代世界和中国物理

科学发展的全貌，是一部目前国内为数不多的兼具学术价值和阅读乐趣的经典物理丛书。

《中外物理学精品书系》另一个突出特点是，在把西方物理的精华要义“请进来”的同时，也将我国近现代物理的优秀成果“送出去”。物理学科在世界范围内的重要性不言而喻，引进和翻译世界物理的经典著作和前沿动态，可以满足当前国内物理教学和科研工作的迫切需求。另一方面，改革开放几十年来，我国的物理学研究取得了长足发展，一大批具有较高学术价值的著作相继问世。这套丛书首次将一些中国物理学者的优秀论著以英文版的形式直接推向国际相关研究的主流领域，使世界对中国物理学的过去和现状有更多的深入了解，不仅充分展示出中国物理学研究和积累的“硬实力”，也向世界主动传播我国科技文化领域不断创新的“软实力”，对全面提升中国科学、教育和文化领域的国际形象起到重要的促进作用。

值得一提的是，《中外物理学精品书系》还对中国近现代物理学科的经典著作进行了全面收录。20 世纪以来，中国物理界诞生了很多经典作品，但当时大都分散出版，如今很多代表性的作品已经淹没在浩瀚的图书海洋中，读者们对这些论著也都是“只闻其声，未见其真”。该书系的编者们在这方面下了很大工夫，对中国物理学科不同时期、不同分支的经典著作进行了系统的整理和收录。这项工作具有非常重要的学术意义和社会价值，不仅可以很好地保护和传承我国物理学的经典文献，充分发挥其应有的传世育人的作用，更能使广大物理学人和青年学子切身体会我国物理学研究的发展脉络和优良传统，真正领悟到老一辈科学家严谨求实、追求卓越、博大精深的治学之美。

温家宝总理在 2006 年中国科学技术大会上指出，“加强基础研究是提升国家创新能力、积累智力资本的重要途径，是我国跻身世界科技强国的必要条件”。中国的发展在于创新，而基础研究正是一切创新的根本和源泉。我相信，这套《中外物理学精品书系》的出版，不仅可以使所有热爱和研究物理学的人们从中获取思维的启迪、智力的挑战和阅读的乐趣，也将进一步推动其他相关基础科学更好更快地发展，为我国今后的科技创新和社会进步做出应有的贡献。

《中外物理学精品书系》编委会 主任
中国科学院院士，北京大学教授
王恩哥
2010 年 5 月于燕园

Preface

When deciding how to organize a book on surface plasmons, it seemed natural to consider the dimensionality of the surfaces on which they exist. On planar surfaces, which include both semi-infinite surfaces as well as multilayer thin films, there is a rich body of phenomena related to propagating surface plasmons. The same is true for surfaces of nanoparticles having a rich variety of phenomena for localized surface plasmons. Surfaces of nanowires and nanogrooves lie in between these two regimes, and these surfaces support both propagating and nonpropagating surface plasmons. In this book, therefore, we have initially categorized the chapters by surface dimensionality, trying to point out both the differences and similarities of the surface plasmon phenomena in these three regimes.

This book does not hesitate to include mathematical derivations of the equations that describe the basic surface-plasmon properties. After all, it was our desire to base the book on *Mathematica* precisely so that these equations could be explored in detail. Our derivations of the properties of surface plasmons are based on Maxwell's equations in SI units. In Chapter 2, Maxwell's equations are introduced for dense media, i.e., media which can be described by frequency dependent permittivity, permeability and conductivity. Because interfaces are essential to surface plasmons, the electromagnetic boundary conditions are required. Practically all of the results in this book are based on time-harmonic fields that can be most simply represented in complex notation. Unfortunately, in the literature there is no standard definition for the complex functional dependence on time of the electric and magnetic fields. We choose a time dependence of $\exp(-i\omega t)$, which has the advantage of making both real and imaginary parts of the complex optical refractive indices positive numbers as they are generally given in standard handbooks. Other properties of waves, including their group velocity, phase velocity, impedance and Poynting vectors are also derived in Chapter 2.

At optical frequencies (near IR and visible) it has been standard practice until recently to automatically set the permeability equal to unity. With the discovery of metamaterials and the predictions of potentially amazing properties like perfect lenses and invisibility cloaks, it is no longer adequate or safe to do so. In Chapters 2 to 7, the physics of surface waves propagating along single and double interfaces are carefully examined for all combinations of materials with both positive and negative permittivity and positive and negative permeability. As a result, unfamiliar modes such as surface magnons, which depend upon negative permeability, are analyzed in addition to those of surface plasmons. It transpires that it is important to define the refractive index of a medium, n, as the product of the square roots of the relative permittivity and permeability, $\sqrt{\epsilon_r}\sqrt{\mu_r}$, rather than the square root of their product. A new formalism is presented in which the media of single- and double-interface structures are characterized in terms of an ϵ_r'–μ_r' parameter space, represented as a vector in polar coordinates, where the prime denotes the real part. This formalism also uses a medium with a double positive set (ϵ_r', μ_r') to generate the other three sets of media, (ϵ_r', $-\mu_r'$), ($-\epsilon_r'$, μ_r') and ($-\epsilon_r'$, $-\mu_r'$). The properties of guided modes propagating along single- and double-interface structures, obtained by using this formalism, are then discussed in detail in these chapters. With the single- and double-interface model, it is also straightforward to understand the manner in which prism coupling via attenuated total reflection is used to launch surface plasmons on metallic surfaces and what effect the prism has upon the properties of the surface plasmon, such as propagation distance and line width.

In the remaining chapters, the discussion is narrowed to surface plasmons alone (positive μ_r), both propagating and localized modes. Quasi-one-dimensional surfaces, nanowires and nanogrooves, are discussed in Chapter 8 and quasi-zero-dimensional surfaces, nanoparticles and nanovoids, are discussed in Chapter 9. Interactions among neighboring nanoparticles are also considered. Although the Otto and Kretschmann prism-coupling configurations were analyzed in Chapter 2, they are briefly reconsidered and compared to other techniques for launching surface plasmons in Chapter 10. In particular, the Chandezon technique for computing vector diffraction in a semi-analytical way is implemented to discuss the ability of gratings to couple optical energy into surface plasmons. A detailed analysis of this technique is described in the online supplemental materials for this book found at the web site www.cambridge.org/9780521767170. Newer techniques, that make use of near-field interactions to excite surface plasmons, are also described.

The text would not be complete without a discussion of plasmonic materials. There are relatively few metals that are plasmonic at optical frequencies and it is not surprising that both gold and silver are so frequently used in surface-plasmon calculations and devices. The relationship between the complex permittivity of a material and its ability to exhibit surface-plasmon phenomena is considered in Chapter 11. The Drude dielectric function, as a phenomenological model for metals, is also considered in this chapter. Chapter 12 is a survey of various actual and potential applications of surface plasmons. This marvelous effect has already proven itself in the form of label-free biosensing for pharmaceutical development and medical diagnostics. It may soon find even larger applications in nanophotonics and magnetic data storage.

The finite difference time domain (FDTD) technique – a numerical method for computing the response of materials to incident electromagnetic fields when the geometry is too complex for analytical techniques – is described in the Appendix. Although FDTD is not implemented within *Mathematica* (it would take forever to run even simple calculations), it has been used to model some of the examples that are considered within the text and it is shown to deliver highly accurate results. A short discussion of the connection between the Poynting vector and the local power flow is also included in the Appendix.

Most chapters conclude with several exercises that are meant to stimulate further thought about the properties of surface plasmons that could not be covered in detail in the text, and are well worth the time and effort to study. Generally, the *Mathematica* routines that are included with the online supplementary materials are employed to solve these exercises.

Every chapter also has a reference section. The field of surface plasmons has grown so much over the last two decades that no one text can do an adequate job of covering it. The aim of this book is to provide a sufficient level of understanding of surface-plasmon physics so that the reader can both begin to design his, or her, own research program and also be prepared to tackle the scientific literature on this subject. There are literally thousands of journal articles related to surface plasmons. We have tried to cite many of the more important articles, including some which at this point are several decades old or older, for a more historical context, and these should give the reader a good start in further investigations, but there are also many important articles that we did not include or, unfortunately, overlooked.

This book, which represents the product of many months of collaborative work, was on the whole a very enjoyable experience. Obviously, most of the results described in the text are not original to us. Nevertheless, we have striven to make sure of the accuracy of the equations, derivations, *Mathematica* implementations and descriptions of experimental results, and any errors that remain are solely our responsibility.

We would like to express our appreciation for the kind support and encouragement provided by Seagate Technology during the writing of the book. This book could not have been written without the many contributions of the students, post-docs, collaborators and granting agencies, cited in Dror Sarid's (one of the author's) papers related to short- and long-range surface plasmons. Many thanks are also due to Professor Richard W. Ziolkowski for helpful discussions involving metamaterials, and to Tammy Orr and Juliet A. Hughes for their able help in editing chapters of this book. Bill Challener (one of the authors) would also like to thank several of his colleagues who have shared their expertise with him in both the theory and applications of surface plasmons, including Dr. Edward Gage, Dr. Amit Itagi, Dr. Chubing Peng, Dr. Timothy Rausch and Dr. Zhongping Yang.

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Contents

1
Introduction

In 1952 Pines and Bohm discussed a quantized bulk plasma oscillation of electrons in a metallic solid to explain the energy losses of fast electrons passing through metal foils [1]. They called this excitation a "plasmon." Today these excitations are often called "bulk plasmons" or "volume plasmons" to distinguish them from the topic of this book, namely surface plasmons. Although surface electromagnetic waves were first discussed by Zenneck and Sommerfeld [2, 3], Ritchie was the first person to use the term "surface plasmon" (SP) when in 1957 he extended the work of Pines and Bohm to include the interaction of the plasma oscillations with the surfaces of metal foils [4].

SPs are elementary excitations of solids that go by a variety of names in the technical literature. For simplicity in this book we shall always refer to them as SPs. However, the reader should be aware that the terms "surface plasmon polariton" (SPP) or alternately "plasmon surface polariton" (PSP) are used nearly as frequently as "surface plasmon" and have the advantage of emphasizing the connection of the electronic excitation in the solid to its associated electromagnetic field. SPs are also called "surface plasma waves" (SPWs), "surface plasma oscillations" (SPOs) and "surface electromagnetic waves" (SEWs) in the literature, and as in most other technical fields, the acronyms are used ubiquitously. Other terms related to SPs which we will discuss in the course of this book include "surface plasmon resonance" (SPR), "localized surface plasmons" (LSPs), "long-range surface plasmons" (LRSPs) and of course "short-range surface plasmons" (SRSPs).

There are a variety of simple definitions in the literature for SPs. Many of these are inadequate or incomplete. The "on" suffix emphasizes the fact that SPs have particle-like properties including specific energies and (for propagating modes) momenta, and strictly speaking should be considered in the context of quantum mechanics. In this spirit, one might define a SP as a quantized excitation at the interface between a material with a negative permittivity and free charge carriers (usually a metal) and a material with a positive permittivity which involves a collective oscillation of surface charge and behaves like a particle with a discrete energy and, in the case of propagating SPs, momentum. We will find, however, that most of the important properties of SPs can be satisfactorily described in a classical electromagnetic model, which is all that we will employ in this book. A SP may be defined classically as a fundamental electromagnetic mode of an interface between a material with a negative permittivity and a material with a positive permittivity having a well-defined frequency and which involves electronic surface-charge oscillation. It is, of course, relevant to ask whether or not a classical description of SPs is acceptable. Bohren and Huffman address this question for nanoparticles directly [5]. They state,

"Surface modes in small particles are adequately and economically described in their essentials by simple classical theories. Even, however, in the classical description, quantum mechanics is lurking unobtrusively in the background; but it has all been rolled up into a handy, ready-to-use form: the dielectric function, which contains all the required information about the collective as well as the individual particle excitations. The effect of a boundary, which is, after all, a macroscopic concept, is taken care of by classical electrodynamics."

This statement can be extended to all of the systems we are considering, not just small particles. If the objects supporting SPs are large enough that they can be described by a dielectric function (permittivity), then the classical approach should generally be adequate. This will be the case if the mean free path of the conduction electrons is shorter than the characteristic dimensions of the objects in the SP system. In practice it is found that the bulk dielectric constant accurately describes objects with dimensions down to $\sim$10 nm, and that a size-dependent dielectric constant can be employed for objects with dimensions down to about 1–2 nm [6–8]. For a detailed discussion about size effects of the dielectric function for small metal clusters, see Refs. [9] and [10]. As discussed in the Preface, the equations in this text are derived from Maxwell's equations as expressed in the SI system of units.

This text is based on *Mathematica*. *Mathematica* was not simply used as a word processor for formatting mathematical equations, but was also used to generate numerous figures within the text. The *Mathematica* notebooks, which are included in the online supplementary materials at the web site www.cambridge.org/9780521767170, contain all of the *Mathematica* code, color figures and some additional text. The notebooks can be used to regenerate many of the figures. Moreover, the reader may easily modify parameters in the *Mathematica* notebook code and recompute the figure for perhaps a different wavelength range or different material, etc. In chapters that discuss material properties, the refractive indices for a wide variety of plasmonic, noble and transition metals are available for calculations in addition to those materials which are specifically used in the figures. Some examples of the algorithms that are included in the *Mathematica* notebooks are a simple theory of the interaction of light with cylindrical nanowires and nanotubes in Chapter 8, Mie theory for calculations with spherical nanoparticles and nanoshells in Chapter 9, and the theory of Chandezon for vector diffraction of light from gratings in Chapter 10. In general, the reader should open the *Mathematica* notebook for the chapter of interest (it is, of course, necessary to purchase and install *Mathematica* first) and at the very beginning of each notebook there is a section labelled "Code." The experienced *Mathematica* user knows to double click on the downward arrow of the rightmost bracket of this section in order to expand it. The first paragraph in the Code section describes the steps that the *Mathematica* user should employ to reproduce a figure in the text. The reader is strongly encouraged to take advantage of these *Mathematica* features to gain the full benefit of the text! The online supplementary materials also include a pdf version of the color figures and a description of the Chandezon vector diffraction theory.

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