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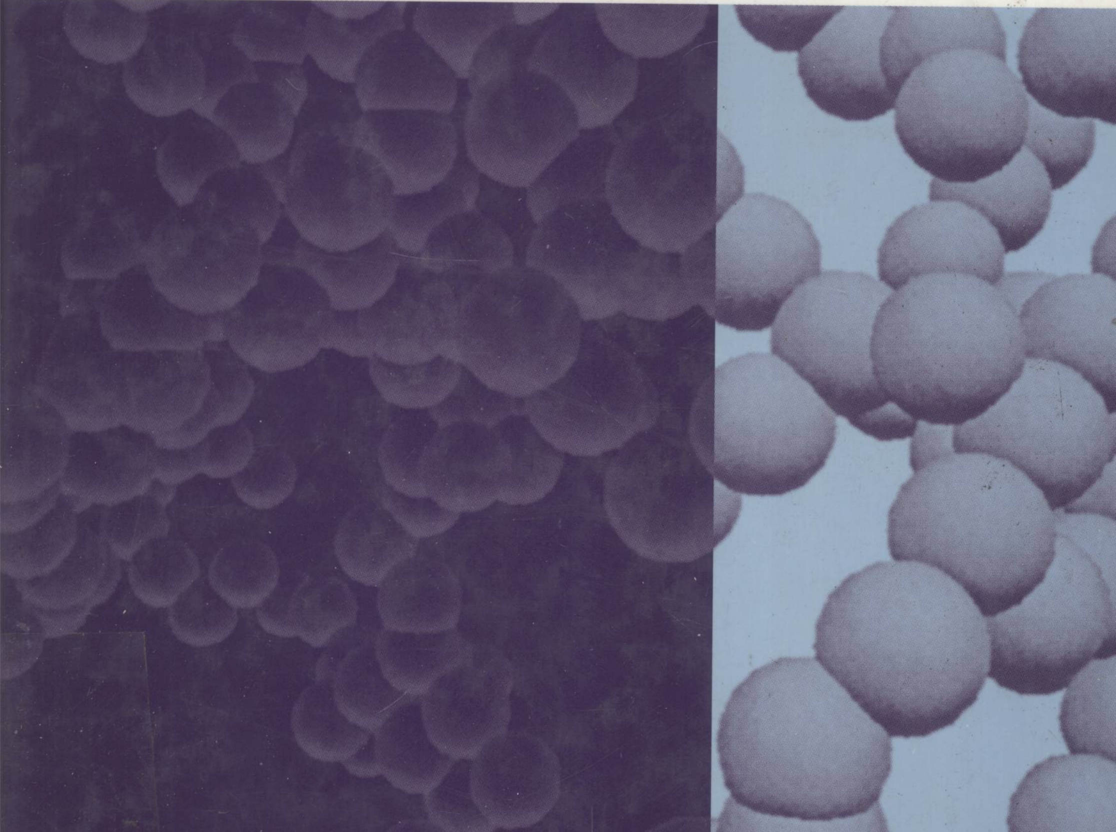
Optics of Nanostructured Materials

Vadim A. Markel
Thomas F. George



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OPTICS OF NANOSTRUCTURED MATERIALS

Edited by

Vadim A. Markel
Washington University, St. Louis

Thomas F. George
University of Wisconsin-Stevens Point

华南理工大学
图书馆藏书

E200100607

A WILEY-INTERSCIENCE PUBLICATION

JOHN WILEY & SONS, INC.

New York • Chichester • Weinheim • Brisbane • Singapore • Toronto

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Published simultaneously in Canada.

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Library of Congress Cataloging-in-Publication Data:

Markel', V. A. (Vadim Arkad'evich)
Optics of nanostructured materials / Vadim Markel, Thomas George,
p. cm.—(Wiley series in lasers and applications)
ISBN 0-471-34968-2 (cloth)
1. Nanostructure mateirals—Optical properties. 2. Optoelectronics—Materials.
3. Nanowires. I. George, Thomas F., 1947- II. Title. III. Series.

TA418.9.N35 M517 2000
621.36–dc21 00-025715

Printed in the United States of America.

10 9 8 7 6 5 4 3 2 1

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Optics of Nanostructured Materials
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PREFACE

One of the most important features of nanostructures, from the point of view of an optical scientist, is that they have characteristic length scales much larger than atomic sizes but small compared to the wavelength of light. In a sense, nanomaterials can be thought of as an intermediate phase between bulk samples, which have characteristic sizes much larger than the wavelength in question, and individual atoms and molecules. The vast majority of optical phenomena involving macroscopic samples can be understood from the purely classical description based on Maxwell's equations. Of course, this statement is not entirely true, since the optical constants of matter are treated as phenomenological while, in fact, they must be calculated microscopically; nevertheless, it is possible to understand the physical optics of macroscopic continuous media from a purely classical point of view. By contrast, the interaction of light with atoms and molecules is generally described only by quantum mechanics. In the case of nanostructures, both approaches are used, which is illustrated by the selection of chapters in this volume. As a rough rule, the quantum description is more appropriate when the optically active electrons (or holes) are free but confined in nanostructured potentials, as in the case of quantum dots and wires (Chapters 11 and 12).

Although there exists a vast literature devoted to the fabrication of nanostructured materials and their physical and chemical properties and applications, very few books, apart from the literature on photonic band crystals [1–4], focus specifically on optical properties [5–10]. Even less attention is paid to the *classical* description of the optical properties of nanostructures. This volume is an attempt to fill in these blanks. Although far from being exhaustive, which is, perhaps, impossible in such a rapidly developing and vast field as physics, it collects under one cover review chapters written by physicists who actively work in the field of optical properties of nanostructured objects, including materials that are not designed or fabricated on purpose (such as carbonaceous soot).

Due to volume limitations, we have had to restrict the scope of this book, and, as a result, it tends to naturally tilt toward the scientific interests of the editors. Therefore, a relatively large number of chapters is devoted to the classical description of *disordered* nanostructured systems, including fractals, nanocomposites, and random metal–dielectric fields. However, several chapters deal with {ordered} systems. The book opens with two chapters on photonic band crystals and photonic crystal fibers. Chapters 3 and 4 are devoted to near-field optical studies of nanostructures objects. Localization of light in disordered dielectrics and metal–dielectric films is discussed in Chapters 5 and 6. The next two chapters discuss in detail nonlinear optical properties of fractal clusters built from nanometer-sized particles from the experimental (Chapter 7) and theoretical (Chapter 8) points of view. Chaos of dipolar eigenvalues and nonlinear susceptibilities in nanocomposites are also considered in Chapter 8. Chapters 9 and 10, devoted to the optics of fractal carbonaceous soot, are closely related to the previous chapters (Chapters 5–8) in the way the electromagnetic interaction is described. However, they discuss distinctly different objects, in which this interaction is off-resonant, in contrast to resonant interactions described in Chapters 6–8. Finally, the two closing chapters (Chapters 11 and 12) review the physics and applications of quantum wires and quantum dots.

We would like to express our deep appreciation of the time and effort that the authors spent in writing their excellent chapters. We are very impressed by the authors' genuine interest in this publication and by their promptness and co-operation. We are also grateful to Maggie Kuhl at the University of Wisconsin—Stevens Point who helped with the editorial work.

VADIM MARKEL

Washington University in St. Louis, St. Louis, MO 63130

THOMAS GEORGE

University of Wisconsin—Stevens Point, Stevens Point, WI 54481

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CONTRIBUTORS

Alexander A. Balandin, Device Research Laboratory, Department of Electrical Engineering, University of California—Los Angeles, Los Angeles, CA 90095

Supriyo Bandyopadhyay, Department of Electrical Engineering, University of Nebraska—Lincoln, Lincoln, NE 68588

Timothy A. Birks, Optoelectronics Group, Department of Physics, University of Bath, Claverton Down, Bath BA2 7AY, UK

Rana Biswas, Ames Laboratory, U.S. Department of Energy, Departments of Physics, and Astronomy and Microelectronics Research Center, Iowa State University, Ames, IA 50011

Sergey I. Bozhevolnyi, Institute of Physics, Aalborg University, DK-9220 Aalborg Øst, Denmark

Yulia E. Danilova, Institute of Automation and Electrometry, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

V. P. Drachev, Insititute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

Thomas F. George, Office of the Chancellor, Departments of Chemistry, and Physics and Astronomy, University of Wisconsin-Stevens Point, Stevens Point, WI 54481

Bernd Hanewinkel, Department of Physics and Material Sciences Center, Philipps-Universität, Renthoff 5, D-35032 Marburg, Germany

Kai-Ming Ho, Ames Laboratory, U.S. Department of Energy, Departments of Physics, and Astronomy and Microelectronics Research Center, Iowa State University, Ames, IA 50011

Alexei A. Kiselev, Physics Research Center, Department of Atmospheric Physics, St. Petersburg State University, Petrodvoretz Ulyanovskaya 1, 198904 St. Petersburg, Russia

Jonathan C. Knight, Optoelectronics Group, Department of Physics, University of Bath, Claverton Down, Bath BA2 7AY, UK

Andreas Knorr, Department of Physics and Material Sciences Center, Philipps-Universität, Renthoff 5, D-35032 Marburg, Germany

Stephen W. Koch, Department of Physics and Material Sciences Center, Philipps-Universität, Renthoff 5, D-35032 Marburg, Germany

Frank L. Madarasz, Center for Applied Optics, University of Alabama in Huntsville, Huntsville, AL 35899

Vadim A. Markel, Department of Electrical Engineering, School of Engineering and Applied Science, Campus Box 1127, Washington University, One Brookings Drive, St. Louis, MO 63130

Eugene F. Mikhailov, Physics Research Center, Department of Atmospheric Physics, St. Petersburg State University, Petrodvoretz Ulyanovskaya 1, 198904 St. Petersburg, Russia

Arkadiusz Orłowski, Instytut Fizyki, Polska Akademia Nauk, Aleja Lotników 32/46, 02-668 Warszawa, Poland

S. V. Perminov, Institute of Semiconductor Physics, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

Marian Rusek, Commissariat à l'Energie Atomique, DSM/DRECAM/SPAM, Centre d'Etudes de Saclay, 91191 Gif-sur-Yvette, France

Philip St. J. Russell, Optoelectronics Group, Department of Physics, University of Bath, Claverton Down, Bath BA2 7AY, UK

Vladimir P. Safonov, Institute of Automation and Electrometry, Siberian Branch of the Russian Academy of Sciences, 630090 Novosibirsk, Russia

Andrey K. Sarychev, Center for Applied Problems of Electrodynamics, 127412 Moscow, Russia

Vladimir M. Shalaev, Department of Physics, New Mexico State University, Las Cruces, NM 88003

Mihail M. Sigalas, Ames Laboratory, U.S. Department of Energy, Departments of Physics, and Astronomy and Microelectronics Research Center, Iowa State University, Ames, IA 50011

Costas M. Soukoulis, Ames Laboratory, U.S. Department of Energy, Departments of Physics, and Astronomy and Microelectronics Research Center, Iowa State University, Ames, IA 50011

Mark I. Stockman, Department of Physics and Astronomy, Georgia State University, Atlanta, GA 30303

Frank Szmulowicz, Air Force Research Laboratory, Materials and Manufacturing Directorate (AFRL/MLPO), Wright Patterson AFB, OH 45433

Peter Thomas, Department of Physics and Material Sciences Center, Philipps-Universität, Renthoff 5, D-35032 Marburg, Germany

Segey S. Vlasenko, Physics Research Center, Department of Atmospheric Physics, St. Petersburg State University, Petrodvoretz Ulyanovskaya 1, 198904 St. Petersburg, Russia

Kang L. Wang, Device Research Laboratory, Department of Electrical Engineering, University of California—Los Angeles, Los Angeles, CA 90095

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CHAPTER ONE

Photonic Crystals

MIHAIL M. SIGALAS, KAI-MING HO, RANA BISWAS, and COSTAS M. SOUKOULIS

Ames Laboratory-US DOE, Departments of Physics and Astronomy,
and Microelectronics Research Center, Iowa State University, Ames IA 50011

1.1. INTRODUCTION

Photonic crystals are a novel class of artificially fabricated structures that have the ability to control and manipulate the propagation of electromagnetic (EM) waves. Properly designed photonic crystals can either prohibit the propagation of light, allow it only in certain frequency regions, or localize light in specified areas. They can be constructed in one, two, and three dimensions (1D, 2D, and 3D) with either dielectric and/or metallic materials.

The concept of photonic band structure arises in analogy to the concept of electronic band structure. Just as electron waves, traveling in the periodic potential of a crystal, are arranged into energy bands separated by band gaps, we expect the analogous phenomenon to occur when EM waves propagate in a medium where the dielectric constant varies periodically in space. There is particular interest in structures that can produce a forbidden frequency gap in which all propagating states are prohibited: Such materials are called photonic band-gap materials and is the topic of intensive studies by many groups theoretically and experimentally [1–4].

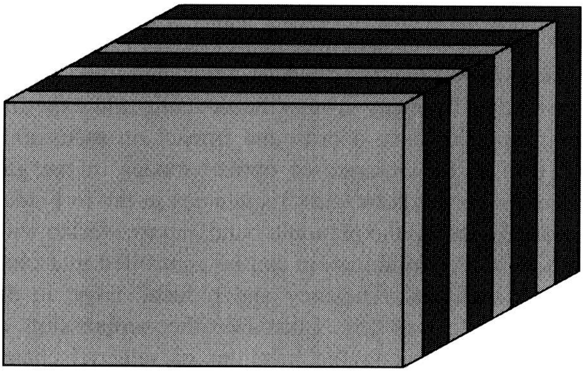
Photonic band gaps can have a profound impact on many areas in pure and applied physics. Due to the absence of optical modes in the gap, spontaneous emission is suppressed for photons with frequencies in the forbidden region. It has been suggested that, by tuning the photonic band gap to overlap with the electronic band edge, electron–hole recombination can be controlled in a photonic band-gap material, leading to enhanced efficiency and reduced noise in the operation of various optoelectronic devices [5]. Likewise, the suppression of spontaneous emission can be used to prolong the lifetimes of selected chemical species in catalytic processes [6–8]. Photonic band-gap materials can also find applications in frequency-selective mirrors, band-pass filters, and resonators. Besides technical applications in various areas, scientists are interested in the possibility of observing

Optics of Nanostructured Materials, Edited by Vadim A. Markel and Thomas F. George
ISBN 0-471-34968-2 Copyright © 2001 by John Wiley & Sons, Inc.

the localization of EM waves by the introduction of defects and disorder in a photonic band-gap material [9–12]. This will be an ideal realization of the phenomenon of localization uncomplicated by many-body effects present in the case of electron localization. Another interesting effect is that, zero-point fluctuations, which are present even in vacuum, are absent for frequencies inside a photonic gap.

There has been a rapid development over the past several years in the fabrication of photonic band-gap materials. Unlike the case of electron waves, which usually have wavelengths on the atomic scale, the wavelengths of EM of interest are several orders of magnitudes larger, varying between hundreds of nanometers for visible light to meters and centimeters for radio- and microwaves. Thus, while for electron waves the periodic lattice is constrained by the crystal structure, the periodic dielectric structures for photonic band-gap materials are artificial structures that can be designed and fabricated to provide a desired electromagnetic response. Therefore, there is a lot of interest in theoretical calculations for these systems and, over the past few years, advances in the field have been characterized by a close collaboration between theorists and experimentalists.

Photonic crystals in 1D have been well known for more than 50 years and are the basis of many devices, such as dielectric mirrors, Fabry–Perot filters, and distributed feedback lasers [13–15]. A 1D photonic crystal is shown in Fig. 1.1(a). It consists of a superlattice of alumina (dielectric constant, $\varepsilon = 9.61$) layers with thickness of 0.4375 mm and air (dielectric constant of 1) layers with thickness of 1.3125 mm. The transmission of EM waves incident on the structure at three different incident angles (0°, 30°, and 60°) is shown in Fig. 1.1. For normal incidence (solid lines in Fig. 1.1), there is a drop of the transmission from 36 up to 77 GHz. The transmission at the center of the gap is almost five orders (almost −50 dB) of magnitude less than the incident wave. The gap is created by the interference of waves due to the periodicity



(a)

Figure 1.1. The transmission for EM waves propagating in a 1D photonic crystal [see panel (a)] for incident angle of 0°, 30°, and 60° (solid, dotted, and dashed lines, respectively). The incident **k** vector is always in the *x,z* plane. Panels (b) and (c) correspond to *E* fields parallel to the *y* axis and in the *x,z* plane, respectively.

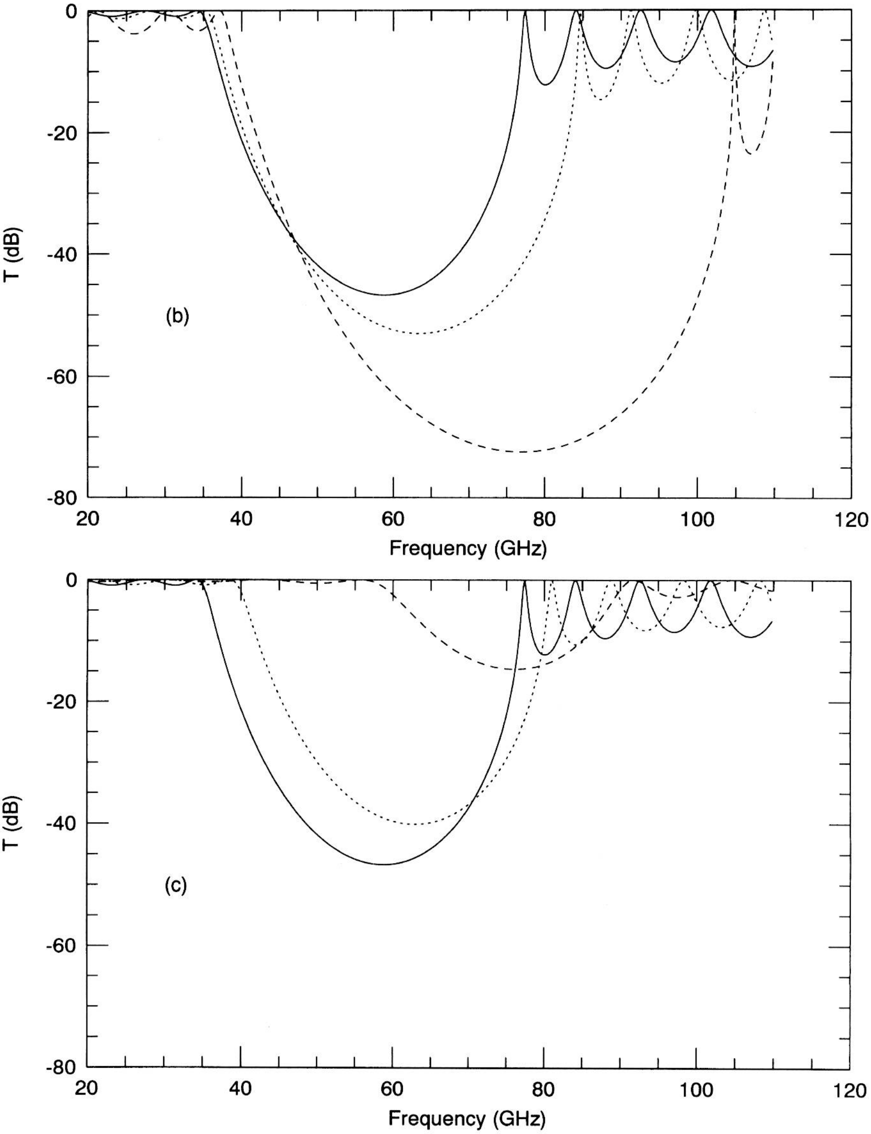


Figure 1.1. (*Continued*)

of the structure along the z axis. One expects that the gap will disappear for other incident angles. Indeed, by increasing the incident angle, the gap increases for the polarization with the electric field out of the plane of incidence *but* the gap tends to disappear for the wave polarized in the plane of incidence.

A realization of a 2D photonic crystal is constructed using infinitely long dielectric cylinders arranged in the so-called 2D triangular lattice [1–4]. The cross-section of this structure is shown in Fig. 1.2(a). The transmission for EM waves with incident **k** vector in the *x,z* plane is shown in Fig. 1.2. We use air cylinders with radius 0.805 mm surrounded by a dielectric with $\varepsilon = 12.25$ (approximately the dielectric constant of GaAs); the distance between the center of the cylinders is 1.75 mm and the total thickness of the system along the *z* direction is 9.1 mm. For waves with the *E* field parallel to the cylinders [Fig. 1.2(b)], there is a small gap at ~48 GHz and a much wider gap at ~80 GHz. As the incident angle increases and the **k** vector is perpendicular to the axis of the cylinders, the second gap moves to smaller frequencies. For the polarization with the *E* field in the *x,z* plane [Fig. 1.2(c)], there is a gap at ~70 GHz for all the angles and for **k** vectors perpendicular to the axis of the cylinders. So, for both polarizations and **k** vectors in the *x,z* plane, there is a gap from 70 to 80 GHz. As in the 1D case, we expect that the gap is going to disappear as the **k** vector moves out of the *x,z* plane since the system is homogeneous along the *y* axis.

It is clear from the previous discussion that we need a 3D structure with periodicity along three directions in order to have a *complete photonic band gap*, that is, a photonic band gap for all the polarizations and all the incident directions. Intense research in the beginning of the 1990s showed that there are some specific structures that poses a *complete photonic band gap* (PBG) [1–4,16,17]. In fact, the first 3D photonic crystal build by Yablonovitch and Gmitter [18] did not have a *complete PBG.* This structure consisted of air spheres embedded in an Al_2O_3

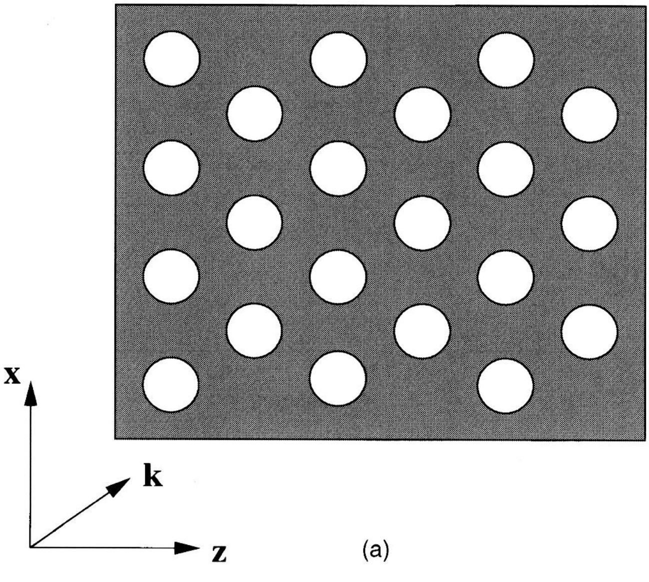


Figure 1.2. The transmission for EM waves propagating in a 2D photonic crystal. The cross-section of the structure is shown in panel (a). The incident angle is 0°, 30°, and 60° (solid, dotted, and dashed lines, respectively). The incident **k** vector is always in the *x,z* plane. Panels (b) and (c) correspond to *E* fields parallel to the *y* axis and in the *x,z* plane, respectively.

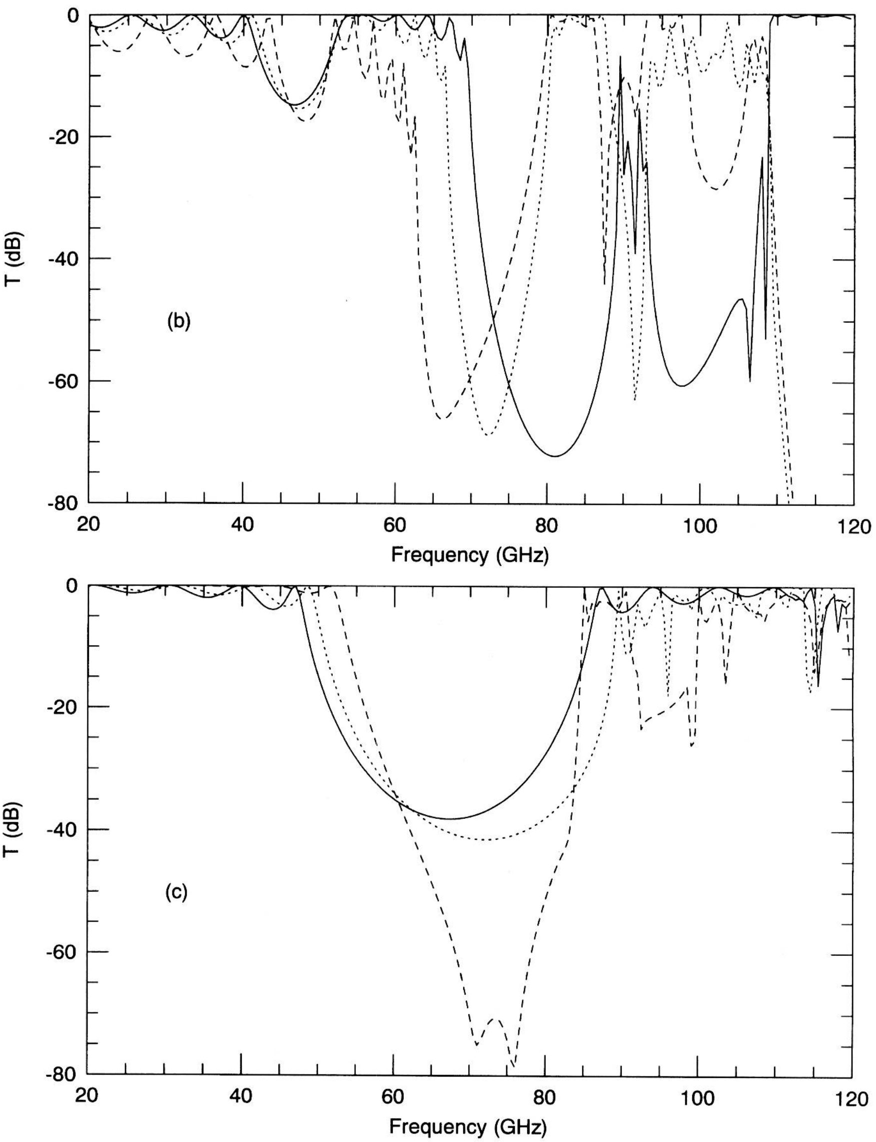


Figure 1.2. (*Continued*)

material forming a face-centered cubic (fcc) lattice. This crystal can be visualized by placing the spheres at the edges and at the center of the faces of a cube. It was constructed by drilling hemispherical cavities on dielectric plates that were stacked together. The whole structure can be constructed by periodically displacing the cube into the space. In contrast to the transmission measurements that showed a *complete PBG* for this structure, subsequent theoretical studies showed that there is not a *complete photonic band gap* for this structure due to a degeneracy of modes at a particular direction [19–21].

More interestingly, Ho et al. [21,22] theoretically proved that the diamond structure consisting of air or dielectric spheres poses a *complete PBG.* A diamond structure is similar to the fcc structure but instead of placing one sphere in each fcc lattice point, we place one more sphere in each lattice point displaced parallel to the body diagonal of the cube by one-quarter of the length of the diagonal. The first photonic crystal with a *complete PBG* was build by Yablonovitch et al. [23]. It has an fcc lattice, but instead of spheres there are cylindrical voids in each lattice point. The so-called "three cylinder structure", can be constructed by drilling holes on the surface of a materials' slab, which are penetrating throughout the whole slab. The holes are forming a triangular array. Three drilling operations are conducted through each hole, 35.26° off normal incidence and spread out 120° on the azimuth. The structure had a *complete PBG* centered at 14 GHz and the forbidden gap width was 19% of its center frequency.

More recently, our group designed and fabricated the layer-by-layer structure shown in Fig. 1.3 [24]. The structure is assembled by stacking layers consisting of parallel rods with a center-to-center separation of a. The rods are rotated by 90° in each successive layer. By starting at any reference layer, the rods of every second neighboring layer are parallel to the reference layer, but shifted by a distance $0.5a$ perpendicular to the rod axes. This results in a stacking sequence that repeats every four layers. This lattice has face-centered tetragonal (fct) lattice symmetry with a

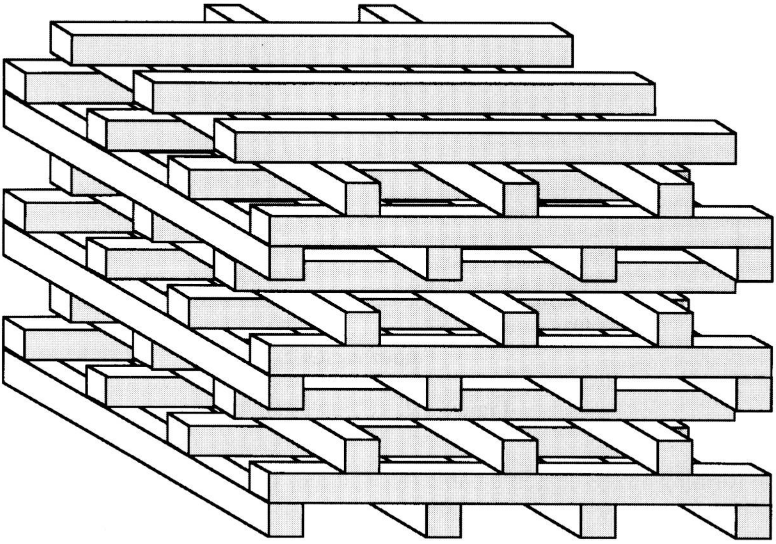


Figure 1.3. Layer-by-layer structure constructed by orderly stacking of dielectric rods. The periodicity is four layers in the stacking direction. Layers in the second neighbor layer are shifted by $a/2$ in the plane.

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