OPTICS OF THIN FILMS

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Optics of Thin Films

(An Optical Multilayer Theory)

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A Wiley-Interscience Publication

JOHN WILEY & SONS

London • New York • Sydney • Toronto

Published in co-edition with SNTL, Publishers of Technical Literature, Prague

Library of Congress Cataloging in Publication Data:

Knittl, Zdeněk.

Optics of thin films.
(Wiley series in pure and applied optics)
"A Wiley-Interscience publication."
1. Thin films—Optical properties. I. Title.

QC176.84.07 K57 530.4'1 73-20896
ISBN 0 471 49531 X

Printed in Czechoslovakia

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Introduction

1.1 Historical and prefatory notes

The anomalous behaviour of matter if it is made to exist in the form of a thin film is an established phenomenon of modern physics which is being exploited both in basic research into the properties of matter and for industrial applications, once the anomalies are under control.

Optics enjoys the historical priority of being the first branch of physics to have been confronted with a thin film anomaly, in the form of coloured reflection from soap bubbles, air wedges between glass surfaces and from fatty patches on water. The earliest descriptions of these phenomena were independently published in the 17th century by several natural philosophers: *Joannes Marcus Marci* (1648), *Robert Boyle* (1663), *F. M. Grimaldi* (1665), *Robert Hooke* (1665). No explanation was given of these colours apart from some vague speculations in terms of reflection and refraction.

Thin film colours did not escape the attention of *Isaac Newton* (1704), yet they eluded an explanation in terms of the corpuscular theory of light. In fact, they remained the touchstone of optics until the beginning of the 19th century, when the foundations of physical optics were laid by *Young* and *Fresnel*.

Interference of light waves was discovered to be the cause of the colours of thin films, and the double-beam treatment of it on a plane-parallel layer has been one of the classical sketches in physics text-books ever since.

In 1833 *G. B. Airy* extended this to a multiple-beam model, but his formulae were unnecessarily exact, and involved, for the thin film phenomena then under study: both the colours of thin films and Newton's rings were adequately described by two-beam interference.

From the theoretical ideas of that time let us also recall *Stokes*' principle of reversion (1849), used to account for the perfect blackness of the central spot in Newton's rings.

However old, all these ideas have kept their importance in modern optical thin film theory. In fact, it was only lack of practical hints that prevented 19th-century physics from developing the concept of the multilayer, and the corresponding methods of analysis. No technical means were known of producing multilayers artificially and no multilayers were registered in nature. The Fabry-Perot étalon of the 1890's was the first element to require the use of exact Airy summation to describe the profile of its Haidinger fringes, but this was still a "silvered" monolayer rather than a real three-layer.

Not until 1917 can one discover a paper concerning a genuine stack of "plates" of alternatively low and high refractivity. The object of the study was not a technical product, but a work of nature. This paper, by no less a person than *Lord Rayleigh*, [1], gave an explanation of the spectral colours observed in the reflection of light on the covers of some coleopterous beetles, known to have laminar structure. We have essentially here a stop-band theory of dielectric multilayers, but, since no artificial stratifications of this kind could be produced at that time, the paper fell into oblivion. Stop-band theory was later redeveloped by various authors in various terms.

The time was hardly ripe then for the application of single layers. An occasional observation (actually an unconscious duplication of earlier findings by *J. Fraunhofer* and by *Lord Rayleigh*) of some atmospherically tarnished lenses led the optics manufacturer *Dennis Taylor* to the elaboration of artificial ageing of glass surfaces by etching, with the effect of reducing unwanted reflection from the refracting boundaries. The interpretation of this effect was in terms of an intermediate refractivity step being introduced between air and the compact glass, as if the layer were acting as a thick one, with no interference involved. Strangely enough, it was noted at the same time that the reflected light changed colour with the thickness etched, and that maximum effect was obtained with a "slatey bronze" tint in reflection.

The British Patent No 29561, granted for this process in 1904, is the first trace of a would-be commercial antireflection technique. A similar patent was granted in 1919 to *L. W. Bugbee* in the United States, the etching being applied during the polishing process. None of these procedures gained a footing, however, because it was not a popular step to admit acids into the process of glass-working.

The real birth of thin film applications in optics had to wait until the middle 1930's, when it was derived from the advances of vacuum technology achieved within the framework of the electronics industry. The vacuum evaporation process in a well-developed plant with an efficient pumping system was discovered to be the best method of producing an interference layer of specified optical properties. *A. Smakula* and *J. Strong* are cited as independent fathers of the single antireflection layer in Germany and the United States, respectively. Parallel to this, *A. H. Pfund* discovered the reflection-increasing potentialities of evaporated high-index layers.

It was subsequently realized that one may evaporate more different layers in succession. This led, in the period 1937 – 1947, to the elaboration of the first multilayer theories for antireflecting and reflecting systems as well as for monochromatic interference filters. *Pierre Rouard, Antonín Vašíček* and *A. W. Crook*, [2-4], may be regarded as the main contributors to the generalization by recurrence of the Airy formulae to multilayer systems, although a number of other authors have made various *ad hoc* computations as well. Important design work of deep foresight, unfortunately confined to patent literature, is due to *Walter Geffcken*, [5], who also pioneered wet and gaseous-reaction deposition processes. *A. G. Vlasov* seems to be the Russian classic of this era, [6].

Exacting technological requirements for the production of optically effective and physically stable multilayers have caused the practical developments to be mostly confined to the laboratory phase. The manufacture of the Fabry-Perot type filter, which spread after 1945, may be regarded as a real commercial hit.

The Airy summation was also used by *S. Tolansky* in 1942 when analysing the Fizeau fringes in a silvered-wedge film.

Post-war development is characterized by increased confidence in the rôle of optical multilayers, with corresponding efforts in the experimental and theoretical spheres. Special evaporation plants were developed for optical multilayers (with only a minority of firms adhering to additive wet processes). Large scale research was undertaken into materials suitable for various spectral regions and satisfying the requirements of economical manufacture and prolonged use of the films. Methods of controlled deposition were developed, optical constants, porosity, structure, adhesion, etc. were studied and an enormous amount of knowledge and skill was put into this new field.

Last, but not least, theory was invoked to discover new possibilities of spectral filtering in the general sense of the word, which after all it the predominant use of interference films in optical applications. Theory was also expected to help in interpreting measurements of optical constants, in setting the production tolerances and in guiding the development of new or modified systems.

Early milestone papers of this second era are, for example, the *Hadley-Dennison* electromagnetic treatment of the reflection and transmission metallic interference filter, [7], the published thesis of *F. Abelès*, [8], being the first general treatment of stratified media in terms of the electromagnetic theory of light, and a paper by *E. T. Welford*, [9], introducing matrix computations different from those of Abelès. Pioneering design work was being undertaken by *F. A. Turner*. The first conference on Thin Films summoned by the Marseille University in 1949 showed how widespread and lively the experimental and theoretical activities actually were at that time, [10].

In the subsequent years four basic text-books on thin films appeared, each having its own method of approach and choice of material, thus supplementing each other in the demonstration that a self-sustained new discipline was being born. These were the works of *H. Mayer*, [11], *O. S. Heavens*, [12], *A. Vašíček*, [13] and the advanced though less accessible treatise by *G. V. Rozenberg*, [22], (to be followed only much later by the now well-known books by *H. Anders*, [23] and *H. A. Macleod*, [24]).

From then on it would hardly be possible to follow the stormy development in any balanced proportions. Reference to literature in the present book is kept at a moderate level, the goal being neither monograph-like completeness, nor a historical look at every stage of the development. At this point the author would like to make an apology to the effect that any shortcomings of documentation should be interpreted as his insufficient absorption of all the existing material rather than anything near discrimination.

In the early stages, use of the theory was prevalently analytical: interesting combinations, mostly the result of straightforward reasoning, were mathematically studied and possibly improved in detail. Vector summation often helped where general theory denied direct insight. Although thin film theory is a more compact piece of science than, for example, geometrical optics, the final formulae to which it eventually leads do not lend themselves to direct analysis in terms of the many design parameters. Recourse was therefore had to large scale numerical analysis, which was facilitated by the happy coincidence of this opening era of multilayers with the advent of automatic computers. No desk-calculator teams had to be formed in analogy to the optical design teams of the past (which also made it easier to integrate the minority of the theorists with experimental teams).

In the course of time real design theory began to grow, developing new concepts and special procedures for various purposes. Analogies with network theory were established, which made it possible to design some analog computers and elaborate exact numerical syntheses. Large computer programs were also written for automatic differential corrections.

The theory of optical interference films has by now developed into a specific chapter of physical optics as regards its principles, but the design aspects have formed it into a clean-cut technical discipline of about the same importance for the science of optical filters as network theory is for electrical filters. The author is tempted to apply a quotation from a network theory classic to the effect that the science "... is a beautiful blend of mathematics, physics and engineering." This book on thin film theory will try to contribute to the substantiation of the analogous belief that there is a beauty in only skin deep.

It should be admitted that the interdependence between theory and practice is much stronger in optics than in electronics, starting with the fact that the thin-film theorist cannot assemble his systems from commercially available separate parts with technical data printed on them. The road to a practical realization of theoretical ideas is more arduous here, which makes some of the practitionists sometimes think that enough has already been done by the theorists to keep technology busy over the next decade or so. This view can be justified if we admit that the discovery of fascinating ideas may be postponed in harmony with the needs of practice. Alternatively, we may tolerate the thriving of some pure theory if the rest of the science is flying at an operational height.

This is the philosophy adopted in the present work which is to be regarded as a text-book meant for advanced university courses or as a reference book for those working in the field. The aim is to acquaint the reader with modern concepts and the procedures of that part of the electromagnetic theory of light which is today applied to stratified media in optical applications. A rough knowledge of Maxwell theory is assumed.

Multilayers are regarded as elements affecting the propagation of infinitely extended plane waves, none of the recent developments of wave guiding inside thin films having been considered.

The evolution of the basic theory is the first objective. The dominant method of approach is matrix treatment, but other useful computational methods are also followed. The now orthodox material on dielectric films is presented in more or less established notations. There was more pedagogic scope for the author in working through the involved formalism of metallic films, providing at the same time for a maximum of insight into the new physical situations occurring there. Some less well known results may be found in the sections dealing with the inner field. The fundamentals of inhomogeneous layers are presented in close connection with the formalism established for homogeneous multilayers.

Wherever possible in the intermediate stages of the presentation, applications are introduced in special chapters written from the view-point of design. It was not the authors ambition to present anything like a complete design theory for thin films, which would call for a special volume, possibly written by several authors. It is hoped though that enough of this material has been incorporated to make the book well balanced between basics and applications. The chapters on thick layers and on perturbations should add to this balance.

A substantial chapter from the other side of the balance is devoted to a type of exact synthesis, which has haunted the author's mind for some years. A number of unpublished results are presented here in full awareness of the fact that their elegance surpasses their utility, at least in the foreseeable future. Nonetheless, the general views obtained here, and the network physical analogies, may deepen the understanding of thin film interference as well as supporting the educational value of the book.

In addition to this, the equivalent of several papers or notes, so far withheld from separate publication, is dispersed over other parts of the book, (particularly in 2.6.5, 3.3, 3.4, 5.3, 6.3.4, 7.1.4, 11.3 and in Ch. 12), it being left for the conversant reader to assess their usefulness in the presentation of the particular subjects.

Finally a few words on how this book came into being. As is often the case, the nucleus was provided by personal notes piling up over the years in industrial research, some of the results having been published in scientific journals. The occasion to lecture on thin films at the university later initiated a process of arrangement and growth resulting in two intermediate stages of condensation: Lecture Notes for the 1966 Czechoslovak Summer School and an enlarged version of these in English for my 1969 stay as guest lecturer at the *Kungliga Tekniska Högskolan*, Stockholm. My thanks here go to Professor E. Ingelstam for his friendly help and encouragement to make the last, but longest, leap between these lecture notes and the present book.

This book could not have been written, had I not had the opportunity