

OPTICS OF THIN FILMS

Z. KNITTL

Optics of Thin Films

(An Optical Multilayer Theory)

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Contents

1. INTRODUCTION	11
1.1 Historical and prefatory notes	11
1.2 The working hypotheses and the practical bearing of the theory	17
1.3 Wave equations of the electromagnetic field	20
1.4 The plane wave in a homogeneous medium	25
1.5 General types of solutions in non-homogeneous media	28
1.6 Transport of energy	29
1.7 Polarization states	32
2. OPTICS OF DIELECTRIC LAYERS	35
2.1 Notation and preparatory remarks	35
2.2 Boundary conditions in admittance notation	37
2.3 Fresnel's formulae	39
2.4 Input admittance at a boundary	40
2.5 Matrix treatment of reflection and transmission	41
2.5.1 Admittance matrix	41
2.5.2 Phase matrix	42
2.5.3 System transfer matrix	42
2.5.4 Evaluating the transfer coefficients from \mathbf{S}	42
2.5.5 Interference matrix and evaluation of the transfer coefficients	45
2.5.6 Reduced matrix work — R-coefficients only	46
2.6 Recursion formulae for multiple reflections	47
2.6.1 Generalized Airy summation	47
2.6.2 An algorithm for complete Airy summation	49
2.6.3 Vector approximation of r	52
2.6.4 Vector approximation of r/t	53
2.6.5 Quality of the approximations	53
2.6.6 Internal transfer coefficients	60

2.7	A recursion pattern for the input admittance. The Smith Chart	62
2.8	Notes on numerical computations	66
3.	BASIC DIELECTRIC DESIGN UNITS	69
3.1	The single layer	69
3.1.1	Amplitude and energy coefficients	69
3.1.2	Analysis of the energy formulae for normal and oblique incidence	70
3.1.3	Reducing and increasing reflection by one layer. Observation in white light	75
3.1.4	The single layer as a prototype of monochromatic filtration	79
3.1.5	Theoretical principles of measuring the optical parameters of thin films	80
3.2	Periodic structures	83
3.2.1	Fundamentals of typology and notations	83
3.2.2	A rough orientation in a HLH. . system	86
3.2.3	Reflection at cardinal points $\varphi = (2l - 1) \pi/2$	88
3.2.4	Latent ($\lambda/2$) layers	92
3.2.5	Use of $\lambda/2$ layers as correcting elements	93
3.2.6	Stop-band theory	96
3.2.7	Symmetry and periodicity	103
3.2.8	Oblique incidence	104
3.2.9	Phase dispersion	107
3.3	Broad-band antireflection by discrete points	109
3.3.1	The antireflection problem	109
3.3.2	Principles of achromatization by discrete points	111
3.3.3	Examples of use	118
3.4	Some detuned multilayers	130
3.4.1	The simulated antireflection bi-layer	131
3.4.2	Design of detuned three-layer antireflection coatings	134
3.4.3	Some further antireflection multilayers of the detuned type	139
3.4.4	Beam-splitters corrected by detuning	141
3.4.5	Detuning as a means of interpolating reflection levels	143
3.4.6	The sloping edge	144
3.5	Applications of periodic structures	148
3.5.1	Interference mirrors. Low-pass and high-pass filters	148
3.5.2	The single-half-wave monochromatic filter	159
3.5.3	The double-half-wave monochromatic filter	162
3.5.4	Notes on multiple cavity filters	173

CONTENTS

4. OPTICS OF METALS	182
4.1 Generalizing the Snell law. The inhomogeneous wave	183
4.2 The invariants and Ketteler's relations	187
4.3 Orientation of the vectors E and H after refraction	189
4.4 Fresnel's formulae on a dielectric/metal boundary	190
4.5 Poynting's vector in the refracted wave	192
4.6 The single metallic film	193
4.6.1 Computing the outer field	195
4.6.2 Computing the inner field	200
5 GENERAL SYSTEMS OF LAYERS	213
5.1 Absorption in a layer system	214
5.2 Potential transmittance and absorptance	218
5.3 Distribution of absorption across a layer system for the case of weakly dissipative media	223
5.4 Total reflection	229
5.5 Frustrated total reflection	233
6. GENERAL THEOREMS ON STRATIFIED MEDIA	240
6.1 Left-and-right-incidence theorem	240
6.2 Reversibility theorem	241
6.2.1 Some conclusions from the reversibility theorem	245
6.3 Equivalence theorems	247
6.3.1 Computing the equivalent parameters for the basic period (0.5B) A (0.5B)	249
6.3.2 Analysing the spectral behaviour of an equivalent layer	251
6.3.3 Stacking the basic period	255
6.3.4 Use of matching layers to manipulate pass-band ripple	257
6.3.5 The stop-band	267
6.4 Theorems concerning induced transmission	270
6.4.1 Front-extension theorem	270
6.4.2 Theorems on maximum potential transmittance	270
7. BASIC METAL/DIELECTRIC DESIGN UNITS	283
7.1 The Fabry-Perot sandwich	283
7.1.1 Basic formulae	285
7.1.2 Fourier expansions for \mathcal{R} and \mathcal{T}	286
7.1.3 The transmission filter	293

7.1.4 The reflection étalon	300
7.1.5 Laser cavity	320
7.2 Dielectric-coated metallic mirrors	323
7.3 Some induced-transmission systems	328
7.3.1 Pohlack's beam-splitters	328
7.3.2 The induced-transmission monochromatic filter	332
8. SPECIFIC COMPUTATIONAL PROCEDURES	337
8.1 Vlasov-Kard recursion formulae	337
8.2 Kard's dual expansions for r/d and $1/d$	339
8.3 Kard's theory of translumination	346
8.4 Circle diagrams	349
8.5 Notes on non-polarizing beam-splitters	359
9. EXACT SYNTHESIS OF TUNED MULTILAYERS	364
9.1 Principles of the procedure	365
9.1.1 Transformation of the formulas	365
9.1.2 Energy relations	368
9.1.3 Finding the a_v , b_v	370
9.1.4 Inversion of the $N_{kv}^{(a)}$, $N_{kv}^{(b)}$	371
9.1.5 Some remarks about the matching methods	374
9.2 The antireflection problem	374
9.2.1 Butterworth and Chebyshev approximations of the low-pass filter	374
9.2.2 Frequency transformations	380
9.2.3 Antireflecting a germanium substrate	380
9.3 The semireflection problem	384
9.3.1 Pohlack's expansion and its Chebyshev matching	385
9.3.2 Tables of synthesized semireflectors	390
9.3.3 Oblique incidence	398
9.3.4 Simplified achromatism by $A_{k2} = 0$	401
9.4 Other cases of matching	405
9.4.1 The sloping edge in Lagrangian approximation	405
9.4.2 The pole-and-zero plot	406
9.5 Phase-change-upon-reflection in terms of the zero distribution	408
9.6 Rational function for $\tan \delta$	410
9.7 Some physical analogies between thin films and electrical networks	412
9.7.1 The transmission-line analogy	412

CONTENTS

9.7.2 The "monochromatic" LC-network analogy	413
9.7.3 The "vanishing film" analogy	416
9.7.4 The rationalized-thin-film analogy	418
9.7.5 Insertion loss—the analogy to optical transmission . . .	421
9.7.6 Darlington's insertion-loss synthesis. Notes on the conditions of realizability	424
10. INHOMOGENEOUS LAYERS	429
10.1 The WKBJ (geometrical optics) approximation	430
10.2 Some exact solutions	433
10.2.1 Exponential layer	433
10.2.2 Linear layer	436
10.2.3 Notes on other solutions. Computer-aided solutions . .	437
10.3 The inhomogeneous layer as part of a layer system	438
10.3.1 Generalizing the Fresnel coefficient for inhomogeneous media in the approximation of geometrical optics . . .	438
10.3.2 The interference matrix of an inhomogeneous film . .	440
10.4 Transfer coefficients of single inhomogeneous layers	444
10.4.1 Exponential layer	445
10.4.2 WKBJ layer	445
10.4.3 Transition layer	446
10.5 A general method of integration	448
10.5.1 Drude's formulae—a first approximation	453
10.6 Approximation by simple reflections. The hyperbolic profile	455
10.6.1 A relationship between the hyperbolic and exponential profiles. The method of simple reflections as a Fourier transform	457
10.6.2 A reciprocity theorem on associated functions	458
10.7 Numerical examples	459
10.8 Notes on the preparation of inhomogeneous layers	471
11. THICK LAYERS	480
11.1 Planeparallel glass blank with coatings	480
11.1.1 A pile of plates	484
11.2 Cemented plate	485
11.3 The cube	487
11.3.1 The simple beam-splitting cube	493

CONTENTS

12. PERTURBATIONS 496

 12.1 Dispersion of dielectrics 496

 12.2 Spurious absorption 507

 12.3 Surface roughness 510

 12.4 Cumulative tint in lenses 517

 12.5 Obliquity effects in lenses 523

 12.6 Production errors 536

INDEX 543

1

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.

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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 practitioners 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