History of Optical Coatings and OSA before 1960

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Introduction

The full history of any scientific subject is impossibly complex, and any account can only be a simplified one. Like other technologies, optical coatings developed over a broad front in many countries with many workers and over a long time. Some discoveries were made and then forgotten and rediscovered later; others were simultaneous but independent. This account is intentionally heavily biased toward The Optical Society, and so, although we will try to retain some breadth in the story, we will concentrate on those workers who were significant in the Society. Others, many of whom we will not mention, were also involved in and made significant contributions to the field.

Beginnings

No one knows exactly when the technology of optical coatings started. As far as optical instruments are concerned, the earliest was probably the simple mirror, and by 2000 B.C. mirrors were common all over the world. Early mirrors were made from anything that could be polished, and their reflectance was simply that of the particular material. Obsidian, jade, bronze, silver, or gold, even pots of water, were all used. The idea of using a coating to improve the reflectance was a later development. We know that the Romans employed many different techniques for mirror manufacture, including some that we can classify as thin films. Glass was a common substrate. Mass production of cheap mirrors involved pouring molten lead over glass, yielding irregular fragments that had somewhat raised reflectance from the lead that stuck to the glass, but quality was generally poor. Better, but more expensive, glass mirrors had films of mercury or gold leaf. Metal mirrors often carried layers of polished tin. Outstandingly clear glass was developed in Murano in the middle of the fifteenth century, and the production of what we would describe as the first modern mirrors followed soon after. These mirrors carried a coating that was primarily a mercury amalgam of tin, although small amounts of other metals were also sometimes added. Thus by the sixteenth century there was a well-established thin film coating industry but the coatings were solely of metals.

The development of interference coatings took rather longer. Of course, nature was first in the use of thin film interference. Color in transparent thin films must have been observed at a very early stage of human development, but it was Isaac Newton who, in the late seventeenth and early eighteenth century, painstakingly established the relationship between film properties and perceived color [1]. He realized that the same effects he saw in his thin films were responsible for many colors in nature and, mistakenly, thought that such effects were responsible for all colors. Not much happened in thin film optics from then until the beginning of the nineteenth century.

Two major events in the early 1800s were the 1802 proposal by Thomas Young that light is a wave [2,3] and the publication in 1810 of Goethe's great book on color [4].

Young was not the first to propose a wave theory for light, and, indeed, for a time the theory was not generally accepted. It took the 1818 work of Fresnel on diffraction [5] to convince the field. The wave theory of light paved the way for the understanding of interference phenomena. Fresnel and Poisson developed the idea of the absentee half-wave and the quarter-wave perfect anti-reflection coating [6]. By the end of the nineteenth century interference in thin films was well understood, had been recognized in nature, and was known to be responsible in the form of tarnish layers for an increase in the transmittance of high-refractive-index lenses.

Goethe's book contained in its first edition a chapter by Seebeck, missing from subsequent editions, dealing with experiments on precipitates of silver chloride where illumination was followed by exhibition of reflection of the very colors used for illumination. Wilhelm Zenker [7] realized that this was an interference phenomenon that could be used in photography and also recognized that half-wave spacing of repeated features should give high reflectance at the corresponding wavelength. Zenker's work was the precursor to the Lippmann emulsion that won Gabriel Lippmann the 1908 Nobel Prize in Physics.

Metallic reflecting coatings had also considerably developed during the nineteenth century. Justus von Liebig's [8,9] development of a wet chemical deposition process for silver in the middle of the century had transformed the production of reflectors of all kinds. Interferometers required beamsplitters with semi-transparent reflectors. Astronomy adopted mirrors constructed from stable glass with silver coatings rather than the older, somewhat unstable, speculum metal. Sputtering was sometimes used, but the general view was that it tended to distort the substrates and so it was not much in favor. Then an important paper by Pohl and Pringsheim in 1912 [10] suggested a vacuum process using what was then called distillation, but nowadays thermal evaporation, for mirror coatings. A great advantage of this method was that with a substrate exhibiting a sufficiently high quality of surface finish, the coatings would immediately form a mirror of equal quality without any further need of polishing.

At the beginning of the twentieth century, thin film applications were largely in photographic emulsions and in metallic reflectors. There was as yet no real need for other kinds of optical coatings. Also, strangely enough, although mirrors were much in demand, there seems to have been no great rush to adopt Pohl and Pringsheim's technique.

Early Efforts

The first volume of the *Journal of The Optical Society of America* appeared in 1917. The second issue (numbered as 2 and 3) contains two papers that would appear of great significance to us today but, from their citations, seem to have received little notice at the time. The first paper, on what we would now describe as an interference optical coating, was by Herbert Ives [11], who modified the treatment of a Lippmann emulsion to produce a narrowband reflecting filter of high efficiency that we would now call a notch filter. In the same issue Otto Stuhlmann [12] described his technique for depositing metallic mirrors and beamsplitters by thermal evaporation from wire sources. Later, in volume 2, Frederick Kollmorgen describes a spinning process for the protection by lacquer of silver films, solving the then current problem of applying a thin, uniform film to protect silver surfaces from tarnishing [13].

In the late 1920s and early 1930s, John Donovan Strong pioneered work in the deposition of optical coatings. 1930 he joined the California Institute of Technology and teamed with Charles Hawley Cartwright to investigate the deposition of an enormous number of metals and dielectrics [14]. By early 1931 Strong had coated, with quartz-protected silver, a 6-in. (15.24-cm) reflector. The following year he replaced the coating with one of aluminum. Aluminum had two great advantages. It strongly reflected the ultraviolet and it had an innate environmental resistance due, Strong was sure, to a film of oxide that naturally formed over the surface.

Meanwhile progress was being made in Germany and France. At the Carl Zeiss company in Jena Alexander (Oleksandr) Smakula [15], of Ukrainian origin, developed an anti-reflection coating for lenses, which for several years remained a close secret and was used primarily for military applications.

Walter Geffcken at the sister company Schott in Jena, around the same time, produced the first narrowband interference filters [16]. Like Smakula's, most of Geffcken's early advances were kept secret. Alfred Thelen's account of Geffcken's work [17] makes fascinating reading. In France, Pierre Rouard, in a 1932 paper [18], described his observation of a significant reduction in internal reflectance at a glass surface induced by an overcoat of a very thin metal layer. He presented his thesis in Paris in 1936 [19], and it included an iterative technique for optical multilayer calculations.

In the United States, John Strong, completely independently of Smakula, realized that his evaporated dielectric coatings could be used as a replacement for the tarnish layers that were known to improve the transparency of glass elements. His paper [20] in the *Journal of The Optical Society of America* was the first account of such coatings to appear in the open literature. Strong coated the lenses of a Leica camera that was probably the first ever to be anti-reflection coated by a vacuum process.

August Hermann Pfund, 1939 Ives Medalist and President of The Optical Society from 1943 to 1945, also made contributions in thin film optics. In 1934 he published [21] an account of a dielectric beamsplitter based on a thermally evaporated film of zinc sulfide for use in interferometers and other systems where transmission was followed by reflection at the same surface. In general, we see gradually increasing interest in thin film interference coatings, much of it, of course, directed toward anti-reflection.

A particularly interesting individual of this era was Katharine Blodgett. In 1920 she became the first woman to be employed as a research scientist by the General Electric Company, where she began working with Irving Langmuir, who won the Nobel Prize for Chemistry in 1932. In 1926, she became the first woman ever to be awarded a Cambridge Ph.D. in physics. She then returned to GE, where she continued the work that Langmuir had started on thin films. In the course of this work she devised anti-reflection coatings for glass. Her Journal publications primarily reported films of barium stearate mixed with stearic acid the acid being removed later by soaking in benzene to leave a barium stearate skeleton behind. Very low reflectances could be obtained in this way. Her 1940 patent [22], however, involved the anti-reflection of soda-lime window glass by adding to it a layer of glass containing a metal such as lead or barium that could then be leached out by acid treatment to leave an etched layer of lower refractive index that acted as an efficient anti-reflection coating and, of course, was environmentally resistant.

In 1937, Arthur Francis Turner and Hawley Cartwright began their ground-breaking research in interference coatings including anti-reflection coatings at MIT. The process they used was vacuum evaporation and the materials for the anti-reflection coatings metallic fluorides, magnesium fluoride being specifically mentioned in claim 6 of their 1940 patent [23]. The publication of this patent induced Germany to publish the Smakula patent that had been kept secret. Turner and Cartwright made other advances in anti-reflection coatings including multilayers. Cartwright described to OSA the advantages of anti-reflection for camera lenses [24], while Miller did the same for the moving-picture community [25]. Then Turner joined Bausch & Lomb in 1939, where he ran the Optical Physics Department until his retirement in 1971.

War Years

Optical instruments of all kinds including binocular telescopes, submarine periscopes, range finders, telescopic gun sights, and aircraft bomb sights were required for World War II. The performance of all of these could be much improved, especially for use at dusk or dawn, by the addition of anti-reflection coatings. All the participants on either side in the war were involved in anti-reflection coatings, yet they were treated everywhere as highly secret.

Richard Denton joined the Frankford Arsenal in early 1942. In 1935 the staff consisted of eleven people. By the end of 1943 the staff numbered 1100. His account of his experiences at the Arsenal [26] paints a vivid picture of the rapid problem solving and innovation that was required by the needs of the conflict. Anti-reflection coatings represented only a part of his responsibilities. Magnesium fluoride had been found most satisfactory, and soon virtually all optics were being coated to improve their transmittance.

Around this time, the importance of heating the substrate during the deposition of the magnesium fluoride anti-reflection coatings was recognized. Cartwright and Strong had included heated substrates during deposition in their investigations at the California Institute of Technology [14] and found the tenacity of silver much improved. Also with Turner he had secured a patent on post-deposition baking of magnesium fluoride [27]. Then Dean Lyon, who had worked on thin films at MIT and now since 1941 was working at the Naval Research Laboratory, "stumbled upon that old idea of heating the elements in a vacuum" [28]. He was eventually awarded a patent for this invention [29]. This process was then used for the remainder of the war. After the war, the Bausch & Lomb company employed the magnesium fluoride process in the production of coated elements, and Lyon sued the company for infringement of his patent in what was a celebrated case at the time and that in 1955 was finally decided in his favor by the United States Second Circuit Court of Appeals.

There was tremendous activity in optical coatings during the war, but little of this appeared in the Society Journal. Frank Jones from the Mellon Institute of the University of Pittsburgh, who had been funded since 1936 by the Bausch & Lomb company to investigate the deterioration of glass surfaces, published with Howard Homer [30] a study of anti-reflection of glass by chemical methods. However, the papers that we would recognize immediately as of fundamental significance in the development of thin film optics were by Mary Banning.

Mary Banning gained her Ph.D. from Johns Hopkins in 1941, and, in the summer of 1941, found herself at the Institute of Optics charged with the creation of an optical thin film laboratory [31]. Faced with such a task nowadays we can turn to the established industry, obtain equipment, and study information in books. She had to start from virtually nothing. Even what methods to use was not clear. She decided on vacuum processes as her primary technique, built and operated the equipment, and published four important papers in the *Journal of The Optical Society of America* [32–35], all of which contain a wealth of practical information and represent very much the foundation on which much of the field was built. One of the papers [34] contains what is still the best and fullest description of the design and construction of an immersed polarizing beamsplitter.

Postwar Years

Now, after the war, the subject expanded rapidly. Part of the reason was the impetus given by the war effort to the field. Many people were involved in optical coating and found it an attractive and rewarding field. But also optics was ready for it. Great improvements could be produced by coating camera lenses. High performance could be obtained from reflecting coatings, avoiding the unpleasantness and unpredictability of the wet chemical processes. Interference filters could be made as easily for one wavelength as another and had enormous energy grasp. Thin film polarizers showed high efficiency without the need for expensive crystals. There were, of course, many military needs, but all of optics was expanding. The chemical industry needed infrared instrumentation, and astronomy needed telescopes and instrumentation and especially narrowband filters for increasing contrast of diffuse nebulosities. Binoculars, photographic cameras, microscoscopes, surveying equipment, and navigational equipment all showed vastly improved performance with anti-reflection coatings.

Optical coatings had developed in Germany during the war, and now the results were being brought back to the United States. In 1946 Howard Tanner, who had been with the U.S. Naval Technical Mission in Europe together with Luther Lockhart, both of the Naval Research Laboratory, published a paper on some of the German anti-reflection coatings. One of the coatings they described in detail was a three-layer one based on a quarter-wave of intermediate index next to the substrate, followed by a half-wave of high index and then finally a quarter-wave of low index. This gives high performance over the visible region. It was further analyzed by Lockhart and Peter King [36], and the idea of the half-wave layer that broadens the anti-reflection performance has since appeared in coating after coating and in many publications and patents.

Accurate calculation of the properties of coatings was of considerable interest, and a good number of the contributions to the Journal at this time were theoretical and concerned optical property calculation. Robert Mooney had two papers in the 1945–1946 volume of the Journal [38]. Antonin

Vasicek, the leading thin film worker in Czechoslovakia, published several theoretical studies [39–42], and Doris Cabellero [43] and Walter Welford [44] also contributed. Most of this work used iterative techniques, but Welford succeeded in putting his method into matrix form. Meanwhile, in France, a young Florin Abelès was gaining his doctorate with a thesis that laid the theoretical foundation of the calculation techniques involving characteristic matrices that we almost universally use for our thin films today [45, 46].

It now becomes difficult to keep track of all that was happening in thin film optics, and we give up completely trying to track all of the significant contributions, even just those to the *Journal of The Optical Society of America.*

Pierre Rouard, in early 1944, had returned as Professor of Physics to Marseille and to optical thin films from a forced two-year absence in Clermont-Ferrand working on acoustics. In 1949 the French Centre National de la Recherche Scientifique, recognizing the tremendous expansion of optical coatings, asked him to organize an international conference on optical coatings in Marseille. This was the first truly international conference devoted entirely to the "Optical Properties of Thin Solid Films." A special July 1950 issue of the *Journal de Physique et le Radium* carried the proceedings in a mixture of English and French. Almost everyone of significance in the field was there. Rouard himself of course, Strong (although now much more in astrophysics), Turner, Heavens, Dufour, Greenland, Ring, Abelès, are just a few of the names. Turner gave a paper [47] describing multilayer anti-reflection coatings, dielectric reflectors, reflection filters, narrowband transmission filters, and frustrated total reflection filters.

At the end of the war, many German scientists were recruited to continue their work in the United States under Operation Paperclip run by the Office of Strategic Services. Two notable ones were Alexander Smakula and Georg Hass. Hass was employed by the United States Army Signal Corps and became director of a significant infrared research activity at Fort Belvoir. He and his group wrote many valuable practical papers dealing with such matters as protection of metallic mirrors and the properties of new coating materials [48–52]. Turner was running his research group at Bausch & Lomb, and he became the recipient of a successful and important series of research contracts for infrared thin film coatings for which Hass was contract monitor. The Fort Belvoir contract reports, long out of print but publicly available at the time, span the period 1950 to 1968 and include anti-reflection coatings, beamsplitters, multiple-cavity filters of many different kinds, and much theory on their designs. Ivan Epstein was working with Turner and was responsible for the ideas of symmetrical periods in filter design that are still used to great effect today [53–55]. There were many other achievements. Turner and Harold Schroeder won a Technical Oscar from the Academy of Motion Picture Arts and Sciences in 1961 for their development of a cold mirror coating for the condenser in movie projectors, much reducing the constant fire risk of the extreme flammability of the film stock. He and Peter Berning [56] introduced the concept of potential transmission and devised the induced transmission filter. More information can be found here [57].

The 1950s marked great progress in optical coatings. Thin films could now be recognized as a discipline with workers who could be described as specialists. Books on the subject began to appear, Herbert Mayer's book on thin films appeared in 1950 [58], followed by Oliver Heavens's in 1955 [59] and Leslie Holland's in 1956 [60]. Then in 1960, Vasicek's book was published [61]. Newcomers to the field now had available excellent compact sources of information for rapid learning.

Astronomers began to be interested in narrowband filters. Many of the nebulosities that they were observing were weak emitters of the hydrogen alpha line at 656.3 nm and were difficult to examine against the broadband light from the night sky. Narrowband filters centered on the hydrogen alpha line were found to improve contrast enormously. The study of solar prominences could also make use of such filters, although for examination of features on the solar disk much narrower filters were required and beyond the ability of the thin film deposition methods available at that time. However, George Dobrowolski showed in 1959 how to manufacture ultra-narrow filters using mica cavities [62].

Contemporary publications show clearly the great barrier to progress that was the volume of calculation necessary in deriving the theoretical performance of an optical coating. The theory had much in common with transmission lines, and Smith Charts were commonly adapted for thin film calculations. Approximate techniques were very popular. Computers existed and were occasionally used—Ivan Epstein was an early user for example—but were cumbersome and not always readily available nor user friendly. Workers in the field tended to use empirical methods, tweaking performance by inspired trial and error in the coating machine. Then in 1958, Philip Baumeister at the University of California at Berkeley [63] showed what might be done in an account of the design of a filter by successive approximations on an IBM 650 computer. This marked the beginnings of the computer-aided design of optical coatings.

By the end of the 1950s we could recognize the modern field of optical coatings. Many companies were producing optical coatings, and there were other companies specializing in the supply of equipment and materials.

Now two very significant events, especially for optical coatings but also for the entire field of optics, occurred. On 4 October 1957 the Soviet Union launched the first artificial earth satellite, *Sputnik 1*, ushering in the Space Age. Then on 16 May 1960, Theodore Maiman achieved successful operation of the first laser. Things were never the same again.

Conclusion

Optics has long reached the stage where optical systems without coatings are unthinkable. Thin film coatings play a variety of roles. In many cases they enable optical components and systems better to perform their function that may be quite different from that of their optical coatings. The anti-reflection coating improves transmission and reduces glare, but the function of the system might be to magnify distant objects. Enabling applications were the main driver for optical coatings in the very early days. Later, with the appearance of the narrowband filter and the thin film polarizer, we begin to see components whose critical performance is purely that of the thin film system, thus extending the role of coatings well beyond that of a purely enabling technology. By 1960 that extension of the role of optical coatings was becoming clear.

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