Optical interference coatings–yesterday and today

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Although they are common in nature and must certainly have been observed by early man, we have to wait till Newton for the first truly scientific study of what we now understand as interference effects in thin films. Young, Fresnel, and Maxwell all contributed and the theory was well established by the beginning of the 20th century. Coatings depending on interference, at this stage, were in their infancy and antireflection and decorative coatings, and color photography were the primary applications. By the middle of the 20th century, the situation had changed completely. Today almost the entire field of optics depends on interference optical coatings. This paper will start with a rapid account of the history and end with a survey of the range of interference coatings that are employed today with a fleeting glimpse of what might be in the future.

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1. Early history

We shall limit the range of the subject to man-made coatings, but we must admit that Nature had already created optical interference structures long before man's appearance on the Earth.

Mirrors are the earliest optical instruments known to man. They were common all over the world, including the Americas, by around 2000 BCE. Most mirrors consisted essentially of a smooth surface that gave whatever reflectance was characteristic of the material. Metal mirrors coated with mercury, which was known from at least 1500 BCE, were probably the first examples of the use of an optical coating. Mirrors were of particular importance to the Romans, their more expensive ones using polished metal surfaces. A cheaper mass production process involved treating glass with molten lead some of which stuck to the glass to give a reflecting surface. The glass would break into irregular fragments but the pieces could be mounted in holders. What we would think of as the first mirrors of modern performance appeared only in the second half of the 15th century with the development of outstandingly clear glass to which was applied a coating consisting of a mercury amalgam with tin. The island of Murano was where the most famous mirror manufacturers were situated. The use of coatings employing interference effects took very much longer and like most scientific advances is multithreaded.

The objective, scientific study of interference colors essentially began with Robert Hooke's (1635–1703) Micrographia, published in 1665[1]. Hooke accurately described his observations of colors in thin sheets of mica and between two glass plates pressed together, but failed to construct any kind of model that would allow prediction of the colors. It was Isaac Newton (1642–1727) who in his Optiks[2], published in its first edition in 1704, accurately related observations of color to film thickness. Newton's measurements were so precise that Thomas Young (1773–1829) some 100 years later could use them accurately to calculate wavelengths. Young correctly proposed that light propagated as a wave and the colors were a consequence of interference[3,4]. It took more than a decade for the idea of light as a wave to take hold and it might have taken very much longer if Augustin Jean Fresnel (1788–1827) had not quite independently shown that the wave nature of light was necessary to explain diffraction. Fresnel and Poisson introduced the idea of multiple beam interference to explain the complete fringe minima associated with halfwave layers and with quarterwavesof index equal to the geometric mean of the indices of the surrounding media[5].

Virtually simultaneously, in his work on environmentally resistant glass, Joseph von Fraunhofer (1787–1826) observed an antireflection effect produced by attacking the surface of an optical component with acid[6] and correctly attributed this to the formation of a layer of lower index over the surface of the glass. It appears that he never developed this further. Optics probably did not need antireflection coatings are that time.

1831 saw the shape of multiple-beam fringes worked out by George Biddell Airy (1801–1892) but practical applications were still lacking. It was another sixty years or so before the major advance of the revolutionary interferometer[7] of Charles Fabry (1867–1945) and Alfred Perot (1863–1925) was published.

Meanwhile, some exciting things were happening in color photography. It began with Goethe's great book on color, first published in 1810[8]. Thomas Johann Seebeck (1770–1831) was a close friend of Goethe and was much influenced by him. He contributed a chapter to the first edition of Goethe's book describing some experiments involving the illumination of silver chloride. When silver chloride was exposed to light, after a time it began to exhibit the color of the light that had been directed onto it. Others repeated the experiments getting essentially the same results. It was thought that the action of the light was to stimulate the production of colored silver salts that somehow were colored to correspond with the stimulus. This correspondence was not thought to be particularly strange. The most effective work was carried out by Alexandre-Edmond Becqerel (1820–1891) who, the father of the discoverer of radioactivity. An emulsion of silver chloride deposited over silvered, polished copper plates gave colored images, but unfortunately no method of fixing them was known and so there was no commercial application. The explanation of the effect had to wait for Wilhelm Zenker (1829–1899), in his self-published book on color photography[9] explained that the standing wave pattern in the emulsion gave to rise to precipitated silver laminae with a half-wave spacing resulting in interference that reflected the very color that produced the effect. Zenker was clearly brilliant, but moved frequently from one subject to another and so never appeared to receive the credit for his ideas that more persistent effort would have secured. Eventually Otto Wiener (1862–1927) gave the definitive proof of the standing waves and Gabriel Lippmann (1845–1921) received the 1908 Nobel Prize in Physics for the emulsion.

In fact, by way of explaining how it was that the technique could reproduce a wide range of colors without one upsetting the reproduction of another Lippmann[10] worked out the beginnings of a theory of interference structures that included some Fourier integral expressions, perhaps the first hint of a much more recent design technique. Then in 1912, Lord Rayleigh published a complete calculation technique for wave propagation in a stratified medium[11]. Included in this paper was an approximate technique that we can recognize as what was later called the vector method and some thoughts on a continuous variation of refractive index leading to a completely recognizable version of the Fourier transform technique nowadays used for rugate design. But there was no suggestion of using it for design. It was simply presented as a calculation tool. Probably design was still a subject far from the minds of workers at that time. In 1917, Rayleigh[12] revisited the calculation concentrating on a regular array of layers. Tucked away in this paper is the deduction that, for highest reflectance at normal incidence, dielectric quarterwaves of alternating indices are indicated, perhaps the first hint of the possibility of design rather than simple calculation.

What may be the earliest deliberately produced thin film multilayer filters were described in a 1917 paper by Herbert Ives[13]. Here we find the description of a filter consisting of many repeats of two layers of different refractive indices and with a halfwave period, what we now term a notch filter. It began as a Lippmann emulsion exposed to monochromatic light with the silver layers bleached to a dielectric after development. The filter reflected strongly essentially monochromatic light with the advantage of angle tuning. In fact, Richard Neuhauss (1855–1915), a Berlin physician specializing in tropical medicine, but also a well-known photographer, had earlier developed a similar process, not for filters but simply with the intention of improving the color rendering of the Lippmann emulsion.

Meanwhile, there was a slowly growing realization that tarnish films, like the ones found by Fraunhofer, could be beneficial in increasing transmittance of glass elements. Taylor had mentioned the effect in his book[14,15]. But although treatments based on acid etches were used to some extent, not a great deal was happening in the mainstream.

Then suddenly in the 1930's, the field started to move rapidly. The reason for this is not completely clear. Advances in vacuum technology meant that vacuum deposition became a viable and versatile technique for thin film deposition. Optics was becoming sufficiently complex for coatings to bring significant improvements in performance. Whatever the reasons, the time was now right. John Strong had in 1932 introduced vacuum deposited aluminum to astronomy but the major topic in optical coatings was the antireflection coating. Strong published the first paper on a vacuum deposited antireflection coating[16]. Not mentioned in his paper, was that he also coated the lenses of a Leica camera, probably the first ever to be antireflection coated by a vacuum process. Following Strong, Charles Hawley Cartwright and Arthur Francis Turner, working together at MIT, described many different vacuum-deposited thin-film antireflection coatings. Then Cartwright and Turner presented a paper, published only in summary, on a reflector consisting of a series of alternate high and low index quarterwaves[17]. This inspired G. L. Dimmick of the RCA Manufacturing Company to create a short-wave pass filter for use in the monitoring of the photographic recording of sound[18].

Unknown to Strong and the others, Alexander Smakula was busy investigating antireflection coatings at the Zeiss Company in Jena but the work was classified as secret because of its military implications. Also in Jena, at the Schott Company, Walter Heinrich Geffcken, began to study multilayer coatings, first antireflecting but later highly reflecting, again in secret. The work on high reflectance was based on the Rayleigh papers and eventually led to edge filter designs[19].

World War II intervened and the improvement in performance due to antireflection coatings meant that almost all military optics was coated. All parties involved in the conflict, on either side, were engaged in this secret effort. By the time the war ended, coatings were an indispensable feature of optical systems. It is difficult to assess the effect of the war on the optical industry. It encouraged production but inhibited the exchange of research results and there were few publications. The flood of publications after the war suggests that it may have actually inhibited progress. Science and technology do not flourish under conditions of secrecy.

By the end of the 1940's, there was a significant world community of optical coating practitioners who met in Marseille for the first international conference dedicated to thin films the results of which were published in 1950 in the Journal de Physique et le Radium. Optical coating was now a subject in its own right.

2. Later history

Now the field of thin-film interference coatings expanded rapidly along with optics. Lenses of all kinds, especially photographic objectives, needed antireflection coatings. The chemical industry needed infrared measurement and analysis instrumentation. Interferometry needed more efficient high reflectance coatings. Cold mirror coatings for the reflecting condensers in cinema projectors were needed to reduce the ever-present fire risk. Narrowband filters improved the contrast of hydrogen alpha emitting nebulosities against the broadband light of the night sky. There were all kinds of applications for the efficient narrowband filters that could be constructed from thin films. These represented just some of the areas that became important for optical coatings.

Then two major events increased the demand for filters enormously. On October 4, 1957, the Soviet Union launched the first artificial earth satellite, Sputnik 1, starting the Space Age. On May 16, 1960, Theodore Maiman achieved successful operation of the first laser. Things were never the same again.

Lasers needed tuned cavities to operate successfully and the tuned cavity needed terminating mirrors of suitably low loss. The quarterwave stack structure was ideal. There were problems. The high power of the lasers was found to damage not only the coatings but most of the optical components involved in the laser system. Resistance to laser damage, its measurement, its understanding, and its improvement has been the constant topic of a major international conference that has met every year since 1969 in Boulder, Colorado, and is known, inevitably, as the Boulder Damage Conference. Space, too, brought its own needs. Apart from the requirements for coatings in a wide range of optical experiments, there was the problem of how to power everything. Conversion of solar flux to electric power through receivers was the answer and the silicon solar cell was the receiver of choice. Efficiency falls as the cell heats up and so increases in infrared emittance were necessary. Also the bombardment by electrons in the newly discovered van Allen belts caused rapid degradation of the cells. A happy solution to both of these problems was the addition of a thin glass plate to the front of the cells. The glass increased the emittance and also acted as a shield against the fast electrons. Unfortunately, the cement used to secure the plates degraded in the ultraviolet light from the Sun and so a longwave pass filter was added both to protect the cement but also to reduce the ultraviolet flux on the solar cell, because conversion was inefficient at these wavelengths and contributed significant heat. Solar cell covers rapidly became a high-volume product.

The thermal evaporation process was the major, almost sole, process for optical coating until the beginning of the 1980's when it was supplemented by the energetic processes. There were stability problems associated with coatings that were eventually traced to moisture adsorption and desorption in the columnar structure of the thermally evaporated films. This columnar structure could be almost completely disrupted by brutal bombardment by energetic ions during deposition. Various energetic processes became popular; ion-assisted deposition and various forms of sputtering penetrated deeply into optical coating production.

Telecommunication had been moving to optical techniques during the 1980's but in the 1990's the Internet became enormously important. Information transfer was now possible at unprecedented speed and volume and the demand naturally spiraled upwards. Single-mode optical fibers that had been thought to offer all the capacity that would ever be needed became inadequate. Signal multiplexing and demultiplexing was necessary, and the narrowband all-dielectric filter that could transmit one communication channel and reflect all others became the multiplexer and demultiplexer of choice. Although other components were proposed and demonstrated they were never able to match the incredibly short time to production of the thin-film interference filter and then in 1995 Takahashi[20] showed how the temperature coefficient of spectral shift for an interference filter could be made virtually zero by the correct choice of substrate. The enthusiasm for telecom spawned a demand bubble that finally burst in the early 2000's. It had an immediately bad effect on the thin-film industry that had responded to the telecom demand by investment in equipment and development of components of constantly improving performance. But it was a financial disaster not a technical one. There had been an enormous advance in technology that was now transferred to other areas of optical coating. The industry did recover and also gained in expertise and confidence.

3. The present

At present, there is an enormous output of what we might term traditional coatings: reflectors, hot mirrors, cold mirrors, antireflection coatings, bandpass filters, dichroics, and so on. However, the energetic processes, ion-assisted deposition and sputtering are now the processes of choice except in those areas where simple thermal evaporation must be used. Also the number of layers employed in coatings has soared. Several hundred layers are not uncommon in demanding applications.

The phenomenon of the surface plasmon resonance is being used in all kinds of applications but especially in the form of a detector for small amounts of materials. The resonance-like dip in p-reflectance in Kretchmann coupling[21] into a plasmon on the outer surface of a high-performance metal layer is well known. The resonance is very narrow in angle and moves with the addition of very small amounts of material to the outer metal surface. The angular movement can be readily measured. A particularly powerful application is in biochemistry where the surface can be treated with proteins chosen to bind with those that are to be detected. This can be used in connection with all kinds of biochemical processes including the detection of specific pathogens[22].

Structured coatings are an important current topic. The moth eye coating, so called because it mimics a structure over the eye of a moth, and which dates back at least to 1973[23], has resurfaced from time to time[24] with differences in the techniques. It has recently found new life in antireflection coatings for plastics[25−28]. These coatings are well beyond what was visualized in the earlier work. They include multilayer coatings with discrete conventional layers supplemented by etched organic layers that form the outermost part of the coating. It is well known that the final step in index at the outer surface of the antireflection coating wields the greatest influence on the reflectance that can be achieved[29,30]. These coatings are intended not just for the traditional lenses but for plastic windows over instrument panels in automobiles and other applications where traditionally antireflection coatings have not been used.

A different kind of structure can be achieved by oblique deposition of optical coatings. Here the columnar structure can be emphasized so that the layers exhibit enhanced birefringence. Various types of polarization manipulating devices can be constructed[31,32]. However it has been demonstrated also that very oblique deposition can result in very-low-index films that exhibit little or no anisotropy[33]. An index as low as 1.05 has been obtained with SiO_2 deposited at 87° incidence[34].

Lithographically produced metallic grids are used as powerful polarizers. They are available commercially and they can be considered as thin films exhibiting strong form birefringence, essentially dielectric and metallic in orthogonal directions. Their transmission of the appropriate polarization mode can be improved further by the introduction of suitable antireflection coatings[35].

There are other applications of regular structures other than polarizers. Ebbesen[36] found that a regular array of very small holes in a metal film could, under certain circumstances, enhance considerably the transmittance of the metal film at normal incidence at a wavelength that varied with the pitch of the array of holes. The reason for the enhanced transmittance was found to be scattering of the incident light into and out of surface plasmons on either side of the metal film[37]. This process minimized the electric field within the metal and so enhanced the transmittance. It is, in fact, rather like an induced transmission filter. It was found that the enhancement does not necessarily depend solely on holes. A series of small protrusions on the surface of the metal can serve as well to scatter into and out of the plasmons[37]. A similar effect has been created by a corrugated metal film[38]. This combination of periodic grating-like structures and thin films shows great promise and may well be one of the important topics of the future.

Composite materials is one of the oldest areas of optical research dating back at least to the early Egyptians but it has recently become of considerable interest. Metal nanoparticles dispersed in a dielectric matrix can still support surface plasmons as can metal nanorods produced by oblique deposition. The surface area of metal is considerably increased in these composite films and this can be used in plasmonic detectors of enhanced sensitivity. Similar nanocomposites are also being used in efficiency enhancement in solar cells. Here the metallic particles are dispersed in solar cells of enhanced sensitivity. The scattering of the light by the nanoparticles increases the optical path length of the light in the absorber material and this both improves the conversion and permits the use of thinner material[39−41].

Progress continues in coatings for ultrafast applications but several major problems remain. There is the problem of monitoring the thicknesses of the layers. The coatings are assemblies of discrete layers and group delay dispersion involves the second derivative of phase with respect to frequency. Derivatives magnify small variations, and vanishingly small variations in phase can become large fluctuations in the second derivative. Small but significant fluctuations in phase can be caused by quite small variations in the front surface reflectance, an effect that would be negligible in other applications. The fluctuations in group delay dispersion are frequently regular and they can modulate the phase of the frequency spectrum of the pulse. Depending on the amplitude of the fluctuations the pulse shape can be corrupted. The phase change on reflection is largely a function of the depth of penetration of the light into the coating. The greater the range of wavelengths and the greater the required group delay dispersion the greater must be the depth of penetration and, of course, the greater the thickness of the coating. Fortunately, the group delay dispersion is additive and so successive reflections at a series of mirrors will yield the sum of the individual dispersions. Much of the recent work on dispersive mirrors has centered on the combination of coatings. A rather clever technique involves two identical mirrors that are tilted at different angles to the pulse so that the peaks of one variation correspond to the troughs of the other[42]. Some recent developments are detailed by Pervak[43].

Then there is the almost completely unexpected new area of study of thermal noise in coatings. This was found to be of major importance in interferometric detection of gravitational waves but can potentially apply to all optical measurements of exceptionally high precision. This is becoming an important subject which is evidenced by the appearance of a completely new book devoted to thermal noise in coatings[44]. Just a few of the mechanisms that are recognized as possibly significant in the book are: Brownian thermal noise, thermoelastic noise, thermorefractive noise, photothermoelastic noise, cosmic ray noise, thermochemical noise, and Stefan-Boltzmann radiation noise. Improvements to the gravitational interferometric mirrors have been made by reducing the internal friction in the tantala layers by doping them with titania[45].

These represent just some of the active areas in the field of interference coatings. What of the future? Prediction is a dangerous and imprecise activity, but there does seem to be a detectable trend towards an increasing importance of composite and structured materials in interference coatings. Also our confidence in the production of more and more complex structures with hundreds of layers will likely inspire us towards still greater complication.

4. Conclusion

Interference coatings have a long history but that does not indicate any tendency towards stagnation. The field continues to develop at an ever increasing rate. Part of this is in response to the expanding needs of optics in general but thin films are not simply supporting servants of optical instrumentation. They also lead the way, in sensitive detectors, in display systems, in anticounterfeiting and decorative applications, in energy saving, a list that goes on and on. Then there are problems to be solved in the thin films themselves. Much of what is happening will be covered in this conference. It is, and has always been, an exciting time in optical interference coatings.

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