SCIENCE AND TECHNOLOGY OF THIN FILM AND PHOTONIC MULTILAYERS: APPLICATIONS IN NEUTRONS, X-RAYS, LASERS TO TERAHERTZ SPECTRAL REGIMES

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Dr. N.K. Sahoo is the recipient of the DAE Homi Bhabha Science & Technology Award for the year 2012

Abstract

Thin film multilayers as 1D photonic crystal has been serving the science world both technologically as well as with fundamental research interest. It’s the tailored photonic band gap that is made use of in the efficient reflection, antireflection and polarization of electromagnetic radiations from terahertz, infrared, optical, X-rays to neutron wavelength regime. The physical basis of these multilayers is to essentially create an appropriate refractive index (or polarizability) contrast between the layers in order to manipulate the amplitude, phase and polarizations of these electromagnetic radiations to serve the ultimate interests. However, each wavelength and energy regime has specific complex issues in designing and developing these multilayers. Moreover, with the emerging novel electromagnetic sources, the challenges are being compounded regularly to appropriately overcome the materials, numerical design, development and characterization issues. The present article is a small attempt to present a glimpse of the multilayer R&D issues and possible solution in the present science and technology context.

Introduction

Presently, the word “thin film” no longer needs a formal introduction or explanation. Thin film technology, simultaneously, is one of the oldest arts as well as one of the ever expanding science and technology [1]. The field have shown remarkable progresses, especially over the past few decades, depicting unparalleled potentials from scientific to commercial domains. It has been projected to be one of the major processing techniques to fabricate electronic, optical, optoelectronic, photonic, spintronics, magnetic data storage devices, sensors, fuel cells, solar cells, communication devices, etc [2]. Departmentally, thin films and multilayer devices have been dominating the high-end scientific and technology researches in various atomic energy programs pertaining to the material sciences, astrophysical sciences, space-telescope technology, solar cell researches, laser spectroscopy, reactor core-viewing optical periscopes and wide range of sensor technologies. The very basis of interplaying and transporting of electromagnetic radiations emanated from conventional discharges, lasers, synchrotrons, neutron sources critically depend on the single and multilayer thin film coatings which control the propagation and manipulation of these radiations from hard X-ray to far infrared wavelengths extending to terahertz regimes [3]. Several laser spectroscopic researches, analytical spectro-chemical techniques and optical communication programs primarily rely on dense wavelength division multiplexing (DWDM) of electromagnetic pulse or CW radiations utilizing multilayer thin film devices matching to the various customized experimental needs.

Conceptually, periodic thin film multilayers belong to the family of 1D photonic crystal, in which there is periodic variation in the dielectric constant or
polarizability and hence in the refractive index [4]. In general, photonic crystal (PCs) families are periodically distributed material structures in one (1D), two (2D), or three spatial directions (3D) that exhibit stop bands or photonic band gaps (PBGs) as shown in layer geometries in Figs.-1(a) & (b). In this regard, 1D photonic crystal geometry as multilayers or super-lattices is the most practicable as well as demanding option to fabricate devices for scientific, commercial and technological applications. Historically, although multilayer films received intensive study over an century, it was when Yablonovitch and John in 1987 joined the tools of classical electromagnetism and solid-state physics, that the concepts of omnidirectional photonic band gaps in 1D to 3D was introduced [5]. This generalization, which inspired the name “photonic crystal,” led to many subsequent developments in their fabrication, theory, and application; from integrated optics to negative refraction to optical fibres that guide light in ambient. As can be seen in the Fig. 2, a multilayer of alternative layered thin film materials as 1-dimensional photonic crystal is the array of 1D lattice points in the reciprocal space. The very basis of this formulation is to create a contrast in the dielectric or optical properties between the successive layers for predictive manipulation of the propagation of electromagnetic radiations that can be extended to 3D geometries as shown in Fig. 3.
Multilayers for Lasers, X-Rays and Neutrons

Multilayer Coatings, as mirrors, anti-reflectors, filters and polarizers, play vital roles in the extended spectral range of electromagnetic radiations starting from Neutron, X-ray, UV-Vis- IR to Terahertz frequency and wavelength regimes [6]. In spite of distinct differences amongst these radiations there are striking similarities in designing guiding thin film based components commonly used to transport these radiations and beams. One of the essential as well as most demanding requirements in guiding these beams has been the high efficient reflection or anti-reflection components or subsystems that often decide the quality of the high-end experiments, instrumentations or performance. The design and development such mirror system become more complex when issues of polarization and high peak flux pulse nature of the radiation are to be tackled. It is interesting to note that irrespective of the diversity amongst the radiations, numerical design formulations used for multilayer interference laser mirrors can be conveniently adapted for neutron and X-ray mirrors only with the appropriate re-formalism of the refractive index and extinction coefficient variables. Historically, there have been very interesting evolutions in the field of multilayer science and technology, especially related to the numerical design and computations. In mid-sixties, Seeley and Smith [7] developed an interesting method of adapting results obtained in the synthesis of lumped electrical circuits for use in multilayer thin-film optical filters. The concept and the analogy of using the lumped electrical network circuit in designing optical multilayer dielectric edge filter or the wavelength multiplexer are depicted in Fig. 4. Such formulations can also be extended to more complex designs like omnidirectional reflecting multilayers [8, 9]. But presently more complex numerical methods are available [10, 11].

Refractive Index: the prime factor in formulating any Optics and Photonics

The propagation of electromagnetic radiation is generally presented according to an optical formalism in which the properties of a medium are described by a fundamental parameter termed as refractive index. Knowledge of the refractive index is sufficient to predict what will happen to all such electromagnetic radiations at an interface; that is to establish the Snell’s laws and to calculate the Fresnel coefficients for reflection and transmission. The laws of propagation of radiation, and in particular the refractive index, depend on the fundamental phenomena involved in the interaction of radiation with matter. The main process of interaction in the visible region of electromagnetic waves, as well as in all other spectral regions, is interaction with the electric dipole component of the wave and the electric properties of the medium. This interaction can come about by the interaction of the wave electric field with the charges in the medium (inductive interaction) or by the interaction of the wave magnetic field with the electric charges and current-carrying conductors (damping).
of the electromagnetic spectrum is the polarization of the molecules in case of an insulator/dielectric and plasmonic in metals. At higher energies as with X-rays, it is generally sufficient to take into account the interactions with the atoms and at the highest X-ray energies only the electrons need be considered in the interaction process. Neutrons interact with the nuclei of the materials, and also have another interaction with the electrons for those atoms which carry a magnetic moment. Since the neutron or X-ray refractive indices of most condensed phases are only slightly less than that of air or vacuum, total external reflection is more commonly observed instead of the total internal reflection experienced with light of optical regime. The critical angle for total reflection is such that the reflectivity of neutrons or X-ray of a given wavelength from a bulk interface is unity at lower glancing angles (ignoring absorption effects) and falls sharply at larger angles. A critical comparison of the above explanations is presented in the Fig. 5.

### Similarities in Multilayers for Lasers, X-Rays and Neutrons

The propagation of neutron de Broglie waves in a potential field is analogous to the propagation of light waves in a medium with a continuously variable refractive index. The potential can be gravitational, magnetic, or nuclear. For example, slow neutrons follow a parabolic path under the effect of gravity as in classical mechanics. Neutrons in a constant magnetic field experience a torque and undergo precession. In a non-uniform magnetic field they experience a force that depends on the relative orientation of the spin and field vectors.

The similarity of the mathematical descriptions for neutron wave and light propagation gives rise to phenomena that are analogous to those of classical optics. In fact, virtually all the well-known classical optical phenomena that are characteristic of light and

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Light</th>
<th>X-Ray</th>
<th>Neutron</th>
</tr>
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<tbody>
<tr>
<td><strong>Refractive Index</strong></td>
<td>( n = (n) + (-ik) )</td>
<td>( n(r) = 1 - \delta(r) + i\beta(r) )</td>
<td>( n_{np}(r) = \sqrt{1 - \frac{V_{np}(r) + \mu B_{np}}{E}} )</td>
</tr>
<tr>
<td><strong>Electro Magnetic</strong></td>
<td>( \hat{n}_E = \sqrt{\varepsilon_r \mu_r} )</td>
<td>( \delta = \frac{\lambda^2}{2\pi} r_n \cdot \rho_n = \frac{\lambda^2}{2\pi} r_n n_x f_x^2(\omega) )</td>
<td>( \beta = \frac{\lambda}{4\pi} \cdot \mu_n )</td>
</tr>
<tr>
<td><strong>Real Part</strong></td>
<td>( n = \frac{1}{\sqrt{2}} \left( \varepsilon_r^2 + \varepsilon_x^2 \right)^{1/2} + \varepsilon_x )</td>
<td>( \beta = \frac{\lambda}{4\pi} \cdot \mu_n )</td>
<td>( \delta = \frac{\lambda^2}{2\pi} \cdot \rho_n )</td>
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<tr>
<td><strong>Imaginary part</strong></td>
<td>( k = \frac{1}{\sqrt{2}} \left( \varepsilon_r^2 + \varepsilon_x^2 \right)^{1/2} - \varepsilon_x )</td>
<td>( \beta = \frac{\lambda}{4\pi} \cdot \mu_n )</td>
<td>( \beta = \frac{\lambda}{4\pi} \cdot \mu_n )</td>
</tr>
<tr>
<td><strong>Critical Angle</strong></td>
<td>( \theta_c = \sin^{-1} \left( \frac{\varepsilon_r \mu_r}{\varepsilon_x \mu_x} \right) )</td>
<td>( \theta_c = \sqrt{2}\delta = \sqrt{\frac{n r \lambda^2 f_x^2(\lambda)}{\pi}} )</td>
<td>( \theta_c = \sqrt{2}\delta = \sqrt{\frac{\lambda^2 b (\rho_n)}{2\pi}} )</td>
</tr>
<tr>
<td><strong>Definition of the Parameters</strong></td>
<td>( n = \text{Real part} )</td>
<td>( \delta = \text{Real part} )</td>
<td>( \delta = \text{Real part} )</td>
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<tr>
<td>( k = \text{Imaginary part} )</td>
<td>( \beta = \text{Imaginary part} )</td>
<td>( \beta = \text{Imaginary part} )</td>
<td>( \beta = \text{Imaginary part} )</td>
</tr>
<tr>
<td>( \varepsilon_r = \text{Dielectric Constant} )</td>
<td>( \mu_r = \text{Magnetic Permeability} )</td>
<td>( \mu_r = \text{Magnetic Moment} )</td>
<td>( \mu_r = \text{Magnetic Moment} )</td>
</tr>
<tr>
<td>( \mu_x = \text{Electron Density} )</td>
<td>( \mu_r = \text{Magnetic Permeability} )</td>
<td>( \mu_r = \text{Magnetic Moment} )</td>
<td>( \mu_r = \text{Magnetic Moment} )</td>
</tr>
<tr>
<td>( \lambda = \text{Atomic Scattering Factor} )</td>
<td></td>
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</tr>
<tr>
<td>( \theta_c = \text{Critical Angle} )</td>
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Fig.5: Formulation of complex refractive index in the optical, X-ray and neutron wavelength regime depicting the dependance of real and imaginary part on the medium as well as on the material parameters.
X-rays have also been demonstrated with neutrons. In geometric optics, there are not only the refraction and reflection of neutrons by materials, but also special properties in magnetic media. In wave optics, there is Bragg diffraction from crystalline materials, and so on, as for X-rays, but there are also other phenomena completely analogous to classical optics. Essentially the mirror or the high reflecting property of the multilayers designed and formulated for all these electromagnetic radiations is closely associated with the achievement of 1D photonic band gap in an appropriate thin film multilayer geometry. As a specific example, a generalized multilayer formulation with an objective of 1D photonic band gap for the laser applications is presented in the Fig. 6.

Mirrors and Super Mirrors for ultra-fast lasers

Current laser technology characteristically has an inverse relationship between pulse duration and bandwidth, requiring any optical component used in short pulse applications to provide the desired response over a large bandwidth. Due to this increased bandwidth, the time domain shape of the ultra-short laser pulse of femto-second ($10^{-15}$ sec) or less is adversely affected by material dispersion. In general, the longer wavelengths of laser light will travel faster than the shorter wavelengths resulting in an unusable signal. More specifically, the dispersion causes phase velocity of the pulse to increase in relation to the group velocity (hence the term Group-Delay-Dispersion GDD). The GDD of the laser amplifying medium, along with other components of the system, must be compensated for, to achieve or maintain the desired pulse shape. Chirped mirrors, a rather recent development in the area of non-linear optics, have provided a solution for these phenomena which utilizes the multilayer formulations of the super mirror designs [12]. Chirped mirrors are now the essential elements in ultrafast optics in order to compensate for positive Group Velocity Dispersion (GVD) suffered by a short pulse.
laser light pulse, which travels through a dispersive medium. Design and deposition of a chirped mirror are more complicated than that of an ordinary multilayer dielectric reflector, because one has to fit not only an amplitude target, but also a phase target. The Fig. 7 presents the layer design for a Double chirped mirror (DCM). An impedance-matching section and an AR-coating on top of the mirror avoid the oscillation in the group delay.

**Mirrors and Super Mirrors for X-rays**

Compton showed that X-ray reflection was governed by the same laws as reflection of light but with different refractive indices depending on the number of electrons per unit volume. The advances in X-ray optics during the last 25 years were facilitated by the availability of bright synchrotron radiation sources, the invention of multilayer x-ray mirrors with high reflectance, and the development of high resolution zone plates. In this context, it is important to note that the multilayer coated mirrors can reflect X-ray beams efficiently at grazing angles up to 10 times larger than the critical angle of a single layer (Fig.8).

The efficiency is also better even at normal incidence, especially for soft X-rays with $\lambda > 25$ Å in a spectral or angular region around the Bragg angle. These properties make them useful as beam deflectors, spectral filters, collimators or focussing elements. They are efficient polarizers for incidence angles around 45°. The imaging performance of mirrors is considerably better at normal incidence than at grazing incidence angles, and optical systems with multilayer mirrors can be used for high resolution imaging as well. Soft x-ray telescopes for imaging of the solar corona have obtained images with about one order of magnitude better resolution than grazing incidence telescopes; the actual resolution (about 0.5 arc sec) is presently not limited by the quality of the optics but by the detectors, the pointing stability of the spacecraft, and the available photon flux. Pictures from the multilayer mirror telescopes on the SOHO and TRACE spacecraft are available on the Internet at http://umbra.nascom.nasa.gov/eit/ and http://vestige.lmsal.com/TRACE/.

**Mirrors & Super Mirrors for Neutrons**

In 1946 Fermi and Zinn first demonstrated the mirror reflection of neutrons (Fermi and Zinn, 1946). Again this follows the same fundamental equations as optical reflectivity but with different refractive indices. Neutrons are scattered by nuclei and the neutron refractive index depends not only on the number of nuclei but also on how strongly they scatter. A quantity known as the “scattering length” that indicates a nucleus’ ability to scatter neutrons may be defined [13]. A neutron refractive index of any material is a function of the scattering length density (SLD) of its constituent nuclei and the neutron wavelength. As with light, total reflection may occur here when neutrons pass from a medium of higher refractive index to one of lower refractive index. Since the neutron refractive indices of most condensed phases are only slightly less than that of air or vacuum, total external reflection is more commonly observed instead of the total internal reflection experienced with light.

The critical angle for total reflection is such that the reflectivity of neutrons of a given wavelength from a bulk interface is unity at lower glancing angles (ignoring absorption effects) and falls sharply at larger angles. Fermi and Zinn had observed the total reflection of thermal neutrons below the critical angle. Since the neutron refractive index, unlike X-rays, is related to its composition and to the scattering lengths of its constituent atoms, it does not follow any systematics as shown in Fig. 9.
Initially, neutron guides were fabricated from glass plates coated on their inside with a thin layer of natural Ni or enriched Ni. In such guides neutrons are transported by total reflection on the inner guide walls, with typical neutron reflection losses of 1% per bounce. Starting about ten years ago, neutron guides equipped with ‘Super-Mirror’ coatings were developed. A typical neutron supermirror used for guiding thermal neutrons consists of typically ~250 bilayers (500 Layers) of Ni/Ti or Co/Ti of varying thickness followed by ~20 anti-reflecting bilayers of Gd/Ti. So the layer geometry and sequence is quite complex as shown in Fig. 10. Experimentally, such mirrors are realized by sophisticated magnetron sputtering process with precise process control formulations [14]. In this case the substrate is the glass with a typical thickness of 200 micron. The total layer thickness of mirror is 7.2 micron and the glass substrate needs to have such super mirror coatings both the side for the desired waveguide applications as depicted above. With the magnetron sputtering system now readily available, a typical Super Mirror deposition process takes 15-20 hours (one side). Such magnetron sputtering system usually has the provision to load 5-15 such substrates (with standard substrate dimensions 141 mm x 254 mm) while carrying out the multilayer coatings as shown in Fig. 11.
Mirrors for Terahertz Regime (T-rays)

The terahertz (THz or $10^{12}$ Hz) is the part of the electromagnetic spectrum (wavelengths typically ranging from 100 µm to 1000 µm), where most molecules have rotational and vibrational absorption modes and therefore is a rich area for molecular spectroscopy, chemical and biological analysis [15]. The frequency range is also termed the far-infrared but the terahertz term was originally used for the region between photonic devices and electronic transit-time devices where few practical sources of radiation were previously available, termed the THz gap (roughly 200 GHz to 10 THz - or 30 µm to 1.5 mm wavelength) (Fig. 12). In the last 5 years this situation has changed significantly and there are many THz sources available with differing power, bandwidth and operating temperatures. The frequency range is particularly interesting for medical and security imaging and spectroscopy as the radiation is non-ionising and hence is far safer than X-ray or gamma camera techniques. However, with terahertz wavelengths ranging from 100 to 1000 µm, the diffraction limit for this long wavelength light means that it is difficult to measure small volume samples. The development of electromagnetic, artificial-lattice structured materials (photonic crystals), termed as metamaterials, has led to the realization of phenomena that cannot be obtained with natural materials

In the terahertz regime, unlike optical region, interactions of both electric and magnetic components of EM radiation with matter need to be considered. Concepts of Metamaterials or Negative Refractive Index Materials [16] arise primarily due to the following mathematical formulation depicting the relationship of refractive index with electric and magnetic permeability factors as follows:

$$n^2 = \left[ \varepsilon, \mu_r \right] \Rightarrow n = \pm \sqrt{\varepsilon, \mu_r} \quad \quad \text{(1)}$$
Where, $\varepsilon_r$ is the relative electrical permittivity, defined as $\varepsilon_r = \varepsilon / \varepsilon_0$ and $\mu_r$ is the relative magnetic permeability, defined as $\mu_r = \mu / \mu_0$. Usually, for conventional materials $+$ sign is taken. But for metamaterials, both $\varepsilon_r$ and $\mu_r$ are simultaneously negative. This only possible by designing a structured multilayer in which the material property and geometry both carry the similar importance. A typical design of a reflecting multilayer using this formulation of metamaterial multilayer is depicted in Figs. 13(a) & (b).

**Conclusion**

The science and technology of multilayers for transporting as well as manipulating electromagnetic radiations has ever expanding domain both with respect to the design formulations and practical realizations. The wavelength domain starting from terahertz, infrared, visible, X-ray to neutron have their specific complex issues and considerations with respect to design concepts, geometries and material choices. In spite of distinct differences amongst these radiations there are striking similarities in designing guiding multilayer thin film based components commonly used to transport these radiations and beams. It is also interesting to note that irrespective of the different radiation-matter interaction processes, numerical design formulations used for multilayer interference laser mirrors can be conveniently adapted for neutron and x-ray mirrors only with the appropriate re-formalism of the refractive index and extinction coefficient variables. Further, the super-mirror design initially visualized for the neutron beams to extend the angular-range of the reflected mirror coating utilizing a graded geometric layered structure has also drawn significant attention for X-ray and ultrafast laser mirror related technologies. However, with increasing demand on quality, precision, efficiency, dimension and long term stability on the multilayer mirrors there are serious issues related to selection of new thin film materials, processes, process-monitoring and characterization techniques. In the Department a strong scientific and technology base has been established to carry out the R&D and develop such state-of-the-art multilayer devices with challenging as well as customized needs of the nuclear energy programs.

**Acknowledgement**

The author would acknowledge the scientific and technical expertise and contributions of various scientists working in the Atomic and Molecular Physics Division (A&MPD) and also various R&D scientists and engineers of other Divisions in the Department collaborating with the multilayer thin film R&D programs of the Division.

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