Module 7: Scattering Techniques

Lecture 41: Mie, Rayleigh, quantum scattering

- Mie Scattering
- Quantum Scattering
- Rayleigh Scattering

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Mie scattering

When the particle diameter is of the order of the wavelength of incident light, the dominant effect is seen to arise from the wave nature of radiation. Quantities affected include wavelength of the scattered light (or frequency shift), changes in intensity with direction, phase, and polarization. An example of the first variety is seen in laser Doppler velocimetry, module 3. Large particles glowing in a sheet of light are utilized in particle image velocimetry (module 3), but will show strong directionality in light scattering as the particle size is reduced. Changes in the attribute of scattered light can be used for the measurement of particle size or particle velocity. An example considered in article 3 studies dispersion of liquid water into droplets, smaller droplets scattering less light when compared to larger ones. Depending on the extent of dispersion of the liquid phase, droplets may coalesce and produce larger ones that scatter strongly. Thus, the intensity pattern is now jointly developed by

- particle sizes created by atomization of the liquid and
- coalescence of droplets in the jet as determined by jet dispersion and hence the inter-drop spacing.

Scattered light under the circumstances is highly directional as sketched in Figure 7.4 below for a single drop of water. In mathematical terms, Mie scattering is entirely predictable from the electromagnetic field equations.

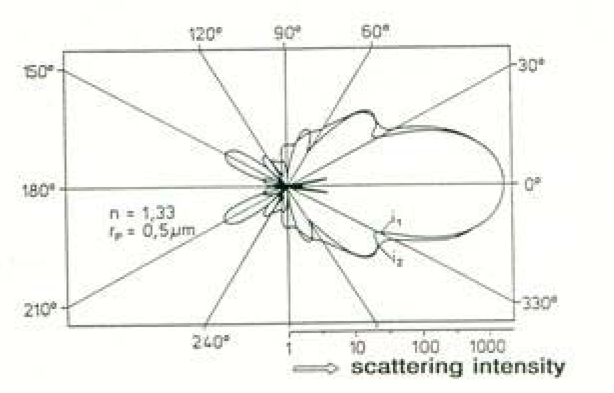

Figure 7.4: Mie scattering from a water droplet indicating a strong dependence of scattered light intensity on angle (from Mayinger, 1994). The polar intensity distribution depends on the shape and texture of the particle and also the ratio of refractive index of the particle with respect to its surrounding medium.

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Quantum scattering

The structure of matter is particulate and comprises atoms and molecules at the fundamental level. Atoms, in turn, have a nucleus with electrons orbiting around. The atom is electrically neutral though internally the positive and the negative charges are spatially segregated. Measurements are related to electronic transitions alone, the nucleus being stable with respect to a majority of external perturbations. When light falls on matter, photons may be absorbed by the medium, raising electrons to a higher energy level. Since the residence time of electrons diminishes with increase in energy, electrons invariably return to their original state, called the ground state . This transition is accompanied by the release of photons of appropriate frequency/wavelength and forms the measurement step in an experiment. The properties of the photon thus released may depend on the properties of the incident radiation and the material properties or any one of them.

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Rayleigh scattering

In Rayleigh scattering , electrons are excited to a *virtual* state below the first energy level above ground (Figure 7.5). Electrons quickly return to the ground state, releasing the absorbed photon. The incident and the emitted photon are identical in terms of energy, and hence wavelength/frequency. The measurement procedure can thus be classified as elastic. Since the incident and the emitted photons are identical, special methods have to be used to distinguish them, by polarization, for example.

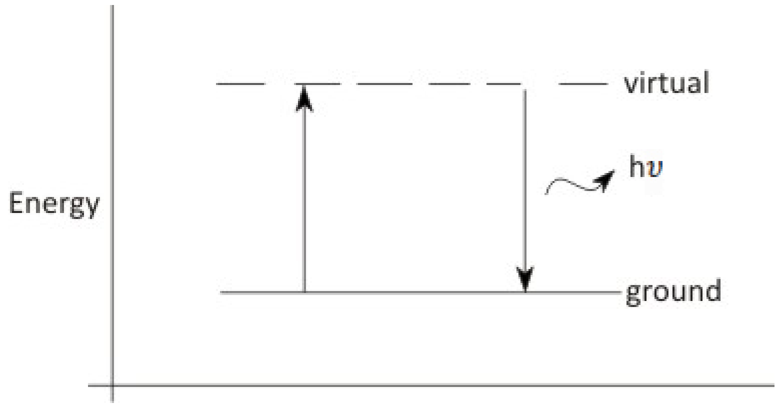

Figure 7.5: Schematic drawing of energy transfer in a Rayleigh scattering process.

The utility of Rayleigh scattering can be seen from the two following results. First, the intensity of light scattered from small particles scales as

$$I = C\frac{d^2}{\lambda^4} \tag{7.4}$$

Here, C is a constant, d is particle diameter, and λ is the wavelength of the incident radiation. Equation 7.5 shows that scattering is biased towards shorter wavelengths, blue in favor of red, for example. It can be used to interpret scattered radiation data from the atmosphere where a large number of atomic-scale nuclei are present. Equation 7.5 can be written in an alternative form

$$\frac{I}{I_0} = C(\lambda; geometry) \times \{N\} \tag{7.5}$$

Here, C is a constant that depends on wavelength and the geometrical arrangement of the source, object, and the receiver. Intensities I and I_0 are scattered and incident values respectively. The symbol N is the number density of the scattering particles. The number density, analogous to the density of the medium, depends on pressure, temperature, and species concentration. Equation 7.5 can be interpreted as a method of finding temperature when all other quantities are prescribed. Alternatively, it is a method of finding the species concentration, for example H_2O in air. See Figure 7.6 and the accompanying article 3 for an example that includes Mie scattered images as well.

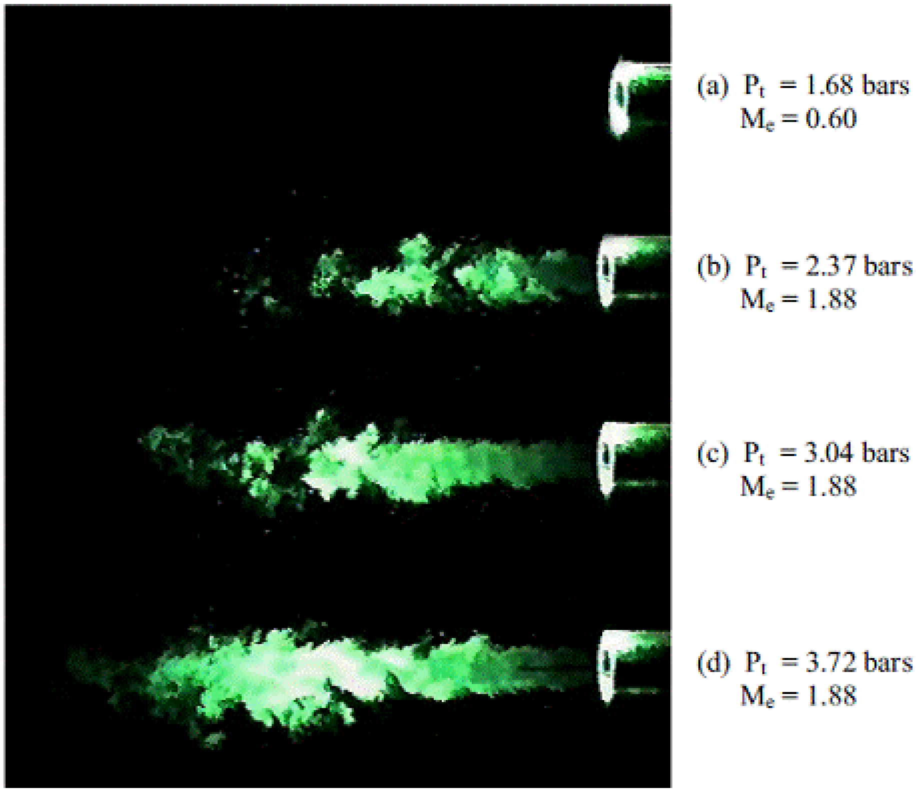

Figure 7.6: Rayleigh scattering images in an over-expanded air jet from a converging-diverging nozzle with water added as a scattering medium (adapted from Kim et al., 1998, article 3).

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Rayleigh scattering (contd...)

Rayleigh scattering is at the interface of wave optics and quantum optics and the properties of the scattered signal can also be determined by the classical electromagnetic field (Maxwell) equations.

When a light beam of strong enough intensity falls on a small portion of a fluid region, the scattered light will contain light intensity fluctuations that are thermal in origin. This effect may not be as pronounced in solids where a lattice structure can create phase differences that result in destructive interference. Phase becomes a near-random variable in liquids and gases, owing to Brownian motion, and a persistent noisy signal is obtained. The statistical properties of the scattered signal, specifically the decay of the autocorrelation function can be related to those in the dielectric constant, and hence the material density. This association is possible because Rayleigh scattering can be explained from a quantum mechanical point-of-view as well as from electromagnetic field theory. Since these fluctuations are thermal in origin, the decay constant of the autocorrelation function will scale with thermal diffusivity of the material. The entire approach is called dynamic light scattering and is useful for thermal diffusivity measurements of fluids under a wide range of conditions. Specific advantages of this route include the small sample size, small experimental time duration, and the absence of any calibration. The method requires detection of small light intensities, adequately resolved in time, and followed by statistical analysis of the light fluctuations (recorded with a high speed detector as a time series of voltages).

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Rayleigh scattering (contd..)

A typical intensity fluctuation recorded in a fluid medium at thermal equilibrium is shown in Figure 7.7. Statistical analysis is possible only for stationary signals for which the time-average is clearly defined (refer module 1 for a longer discussion on signal analysis). Hence, dynamic scattering measurements are to be carried out under constant density (meaning, constant temperature and pressure) conditions, at least for the duration of the experiment. The light intensity signals relate to random density fluctuations. Figure 7.7 also shows the normalized autocorrelation function $g(t)$ of the density fluctuations. In practice, such quantities are obtained by first calculating the Fourier transform of the time series (see module 1). The timescale τ_c associated with the fluctuations can be determined as the intercept of the slope of $g(t)$ with the time axis. Hence

$$\tau_c = \frac{1}{g'(0)}$$

The thermal diffusivity of the liquid medium is then obtained from classical heat conduction (applied to thermal fluctuations) as

$$\alpha = \frac{1}{\tau_c \times q^2} \qquad (7.6)$$

where q is the scattering vector

$$|q| = \frac{4\pi n}{\lambda}\sin\frac{\theta}{2} \qquad (7.7)$$

Here, λ is the wavelength of the incident light, n is the refractive index of the medium, and θ is the angle between the scattering vector and the incident light vector. The refractive index as well as the scattering angle has to be determined as a part of the experiment.

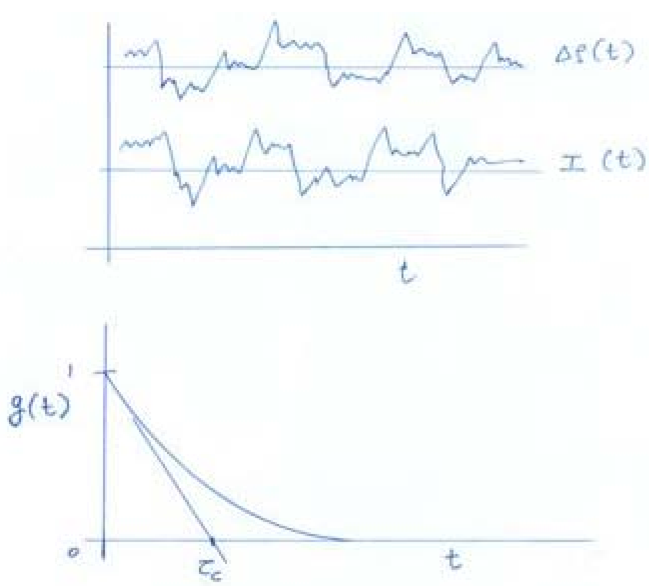

Figure 7.7: Light intensity and density fluctuations recorded during a dynamic light scattering experiment. The graph below is a plot of autocorrelation as a

function of time delay, with τ_c representing the decay rate of the autocorrelation function.

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Rayleigh scattering (contd...)

An important application of Rayleigh scattering is in the measurement of the temperature profile in the middle atmosphere of the earth. The instrument used for this purpose is called LIDAR (light detection and ranging). A schematic drawing of a hand-assembled LIDAR is shown in Figure 7.8. A brief description of the instrument is given here. A powerful pulsed Nd:YAG laser, 10 MW peak power within a pulse width of less than 10 ns, 532 nm wavelength, 30 Hz repetition frequency is used. The high intensity of the laser permits measurements during daytime as much as night-time, not being influenced by the glare of the sun. Scattered radiation is collected in the back-scatter mode with a narrow field-of-view within 0.1 mrad, thus enabling a high signal-to-noise ratio. The telescope aperture is quite large, being around 1.2 m. The laser line is isolated using a Fabry-Perot etalon and a narrow-band interference filter.

The lidar operation can be described as follows. Pulses from the laser transmitter are coupled into the telescope so that, in effect, a collimated beam is transmitted into the sky. Light back-scattered from the atmosphere is collected back by the telescope. The scattered light is collected by the detector through a transmit-receive switch that prevents light from returning to the laser. The switch relies on the change in polarization of the scattered light with respect to the original. Specifically, in the transmit phase, the plane polarized light from the laser passes through the polarizing beam splitter and is converted into circular polarization by a quarter wave plate. The back-scattered light, still circularly polarized, is collected by the receiver, converted into s-polarized light by the quarter wave plate, and directed into the detector by the polarizing beam splitter.

The low altitude signals tend to be strong band and can be removed by a rotating mechanical shutter. The background light at wavelengths different from that of the laser are rejected by using a tiltable Fabry-Perot interferometer (10 GHz band pass) and an interference filter (IF). The signal detector is a photomultiplier tube and a fast counter. Differences in altitude that act as sites of backscatter can be distinguished because of the delay in photons returning to the detector. As per Equation 7.5, the intensity ratio between the outgoing and scattered radiation is a measure of the number density, and indirectly, from the equation of state, the local temperature in the earth's atmosphere.

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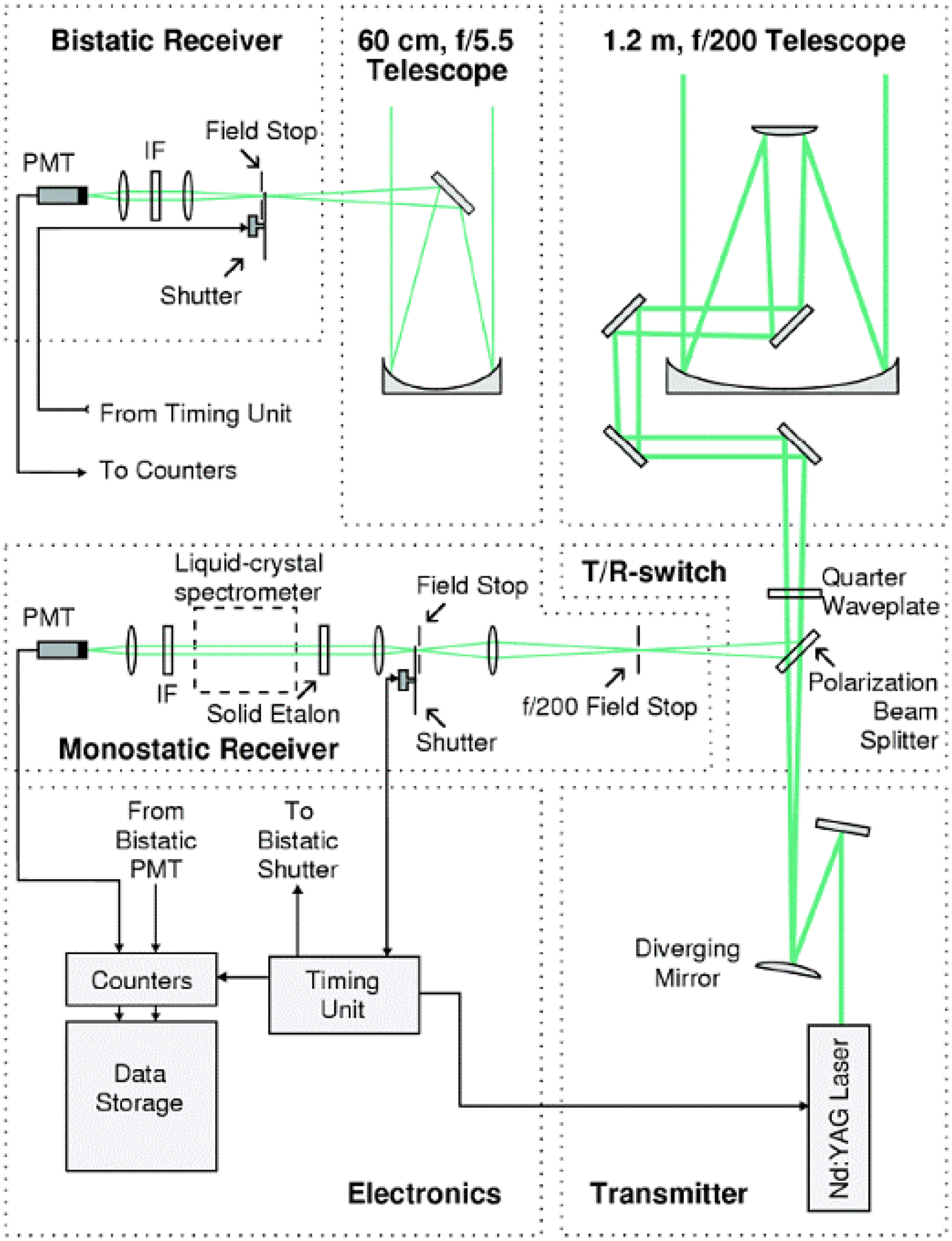

Figure 7.8: Schematic drawing of a LIDAR at the Millstone Hill - MIT Haystack Observatory (after Duck et al. 2001), see article 4 . For applications, see article 5 and article 6 . IF stands for interference filter.

Note on Fabry-Perot etalon :

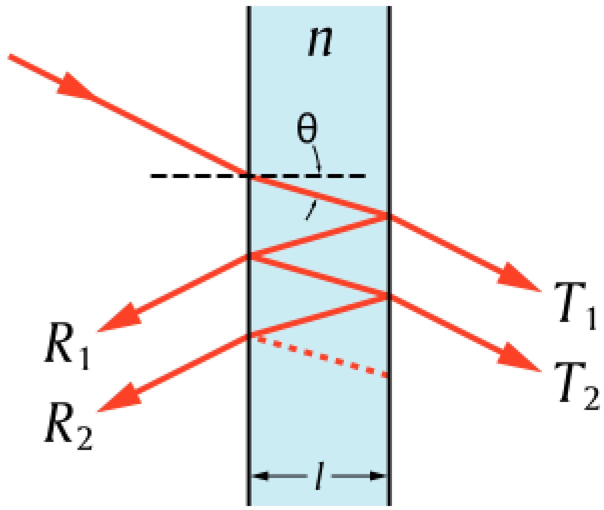

Figure 7.8(I) Schematic drawing of a Fabry-Perot etalon (left); the reflecting surfaces are inside and have a separation of ***l*** . The transmitted light is given a symbol ***T*** while ***R*** is the reflected portion. Figure taken from http://en.wikipedia.org/ wiki/Fabry%E2%80%93P%C3%A9rot_interferometer

The etalon is a pair of partially-silvered mirrors separated by a fixed distance and filled with a medium of known refractive index (Figure 7.8(I)). The silvered portions of the mirrors face each other. The etalon is designed in such a way that the (rays of the) incident light beam will interfere with the (corresponding rays of the) multiply-reflected light beams producing a pattern of constructive interference. Other harmonics are reflected away and the transmitted light beam is at practically at isolated (discrete) wavelengths. In reality, transmittivity of an etalon attains a peak at the designed wavelength and is small at others, but may not strictly reduce to zero. The transmission through an etalon is also a strong function of the reflectivity of the two surfaces. For the configuration shown in Figure 7.8(I), the phase difference between rays T_1 and T_2 can be shown to be

$$\delta = \frac{2\pi}{\lambda} 2nl \cos\theta$$

For reflectance R of the inner surfaces, transmission T_ϵ through the etalon is given by the formula

$$T_\epsilon = \frac{(1-R)^2}{1+R^2-2R\cos\delta}$$

Conditions of maximum transmission for a given wavelength can now be established as a function of wavelength and the angle of incidence. Specifically, for integer values

$$m = \frac{1}{\lambda} 2nl \cos\theta$$

we get

$$\cos\delta = 1$$

Transmission is a maximum under these conditions. In practice, we may have broadened spectrum centered around the desirable value of the wavelength entering the etalon. Passage through the etalon would eliminate most wavelengths and a light beam with sharp spectral peaks would be obtained. Among these, one of them could be most prominent, depending on the transmission function.

An etalon is also used to improve the coherence of a laser. The beam from a laser may appear to have a single color but contains, generally, several frequencies corresponding to the modes of oscillation in the laser resonator. The etalon may be used to separate modes or limit the number of modes. Using interference for mode selection, the desired frequency of oscillation alone passes through. Thus, the coherence of the light output from the etalon is higher than that of the laser.

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Note on interference filter

Figure 7.8(II) Schematic drawing of an interference filter. The dielectric film thickness ***h*** plays an important role in fixing the wavelength that passes through the filter.

The interference filter is fabricated as a thin solid state Fabry Perot interferometer (of thickness *h*) in which the working (reflecting) surfaces are covered by a colored glass plate, Figure 7.8(II). For a given reflectivity, the intensity distribution of the transmitted rays may be treated as having maxima at certain wavelengths given by

$$2nh\cos\theta = m\lambda$$

At normal incidence, $\theta = 0$ and $\cos\theta = 1$. For example, let λ_{-1}correspond to $(m-1)^{th}$ maximum, let $\lambda_0$correspond to m^{th} maximum and λ_{+1}correspond to the $(m+1)^{th}$ maximum of the transmitted rays. Thus

$$2nh = (m-1)\lambda_{-1} = (m+1)\lambda_{+1}$$

The wavelength difference between λ_{-1} and $\lambda_{+1}$is now obtained as

$$\Delta\lambda = \lambda_{-1} - \lambda_{+1} \; and \; nh\;(\lambda_{-1} - \lambda_{+1})/\lambda_{-1}\lambda_{+1} = 1$$ or

$$\Delta\lambda = \lambda_{-1}\lambda_{+1}/nh$$

Assuming the wavelengths to be only slightly different from λ_0 we can write

$$\Delta\lambda = \lambda_0{}^2/nh$$

Clearly, the colored glass filter should be transparent for wavelengths between λ_{-1} and λ_{+1}. A combination of colored glass filter and interferometer will possess the transparency of a Fabry Perot interferometer but will have only one certain interference maximum (at λ_0).

Interference filters can be interpreted as low order Fabry Perot etalons with solid spacers of thickness *h* and refractive index greater than unity. Another difference is that the tuning of the filter is effected by inclining or tilting the interference filter relative to the optic axis of the instrument.

Interference filter using this principle can be obtained by modern vacuum deposition techniques. A thin metallic film is deposited on a substrate by vacuum deposition technique (Figure 7.8(II)). A thin layer of a dielectric material such as cryolite with a known refractive index is deposited over it. This structure is again covered by another metallic film. To protect this film structure from the damage, a glass plate is placed over it. By varying the thickness of the dielectric film, one can filter out all except a particular wavelength.

Interference filters may be designed to pass very narrow bands, but usually sacrifice peak transmittance in that case. A typical interference filter for visible light may have a 7 nm wavelength window with 70% peak transmittance or a narrower window and a lower transmittance. Multilayers can also be designed to transmit or reflect relatively broad bands or reflect infrared while transmitting visible light.

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