University *of Ljubljana*
Faculty *of Mathematics and Physics*

Seminar I$_a$ - 1st year, 2nd cycle

Structural coloration

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Ljubljana, December 2013

Abstract

Structural colors are produced by the interference and scattering, caused by the microscopic structure of a material. In this seminar, selected materials causing structural colors and related mechanisms are presented, including layered materials, photonic crystals, and mechanisms of Rayleigh and Mie scattering. In layered materials with thickness, comparable to the wavelength of visible light, light with distinct wavelengths can be enhanced in distinct directions. Photonic crystals are a generalization of the concept of periodic multilayers, as they have a periodic refractive index in one, two or three dimensions. Both photonic crystals and thin layers produce iridescent color due to the periodicity of their refractive index. On the other hand, Mie and Rayleigh scattering emerge as mechanisms of structural coloration when light interferes with an amorphous structure, which results in non iridescent colors. In structural colors produced by scattering, the angular dependency of scattered light and the color of the material depend strongly on the size of scatterers relative to the wavelength of light. Although examples of structural colors are plentiful in nature, they are only now starting to be used in designed materials. Structural colors are used in industry particularly, because they are very intense and do not fade with time. A wide range of structural colors can be produced by using nanostructures, made from multicomponent materials. Finally, photonic crystals as one of the central structural color materials may also play an important role in the development of optical computers.

Contents

1   Introduction

The origin of colors can be roughly divided into three categories: some colors are due to absorption, others due to emission and the rest are due to the structure of the material - the latter are called structural colors. Typically, the final color of an object is a combination of two or more mechanisms. Examples of structural colors are abundant - the color of the sky and clouds, bright colors of many animals, especially birds and insects, the colors of oil spills and soap bubbles etc. Structural colors are typically very intense and do not fade or change despite being exposed to chemicals or UV light, which makes them very appealing to the industry. They are also environmentally friendlier and less poisonous than pigments. [1]

The term structural colors was first mentioned by Hooke in "Micrographia", which he published in 1665. [2] There he claimed that the color of peacock's feathers was due not to pigments, but due to the structure of feathers - alternating layers of solid material and air. There was no further development until Maxwell's electromagnetic theory in 1873, which could explain interference and diffraction. Later, with the invention of the electron microscope in the early 20th century, it was possible to analyze the structures which produce the bright coloration of numerous animals. [2]

Until recently, the study of structural colors was mainly considered a domain of biology, but has, due to its complexity, now become a lively field of research also in physics and material science. The progress in synthesis of nanostructures lead to the application of structural colors in many fields, such as display technologies, painting, textiles and cosmetics. [2]

This seminar begins with some elementary concepts of our perception of color and explains, how color emerges via mechanisms of absorption and emission. Next, a detailed description of three types of structural coloration is given: thin layer interference, photonic crystals and scattering. The physical background of color, as well as some applications and examples are explained for each type.

2   Colors

In order to understand the origin of colors, we must first know how we perceive them. The human eye has two types of cells that detect light - rods and cones. The rods are much more sensitive than cones and consequently, we use them only in very dim light, when the cones cannot sense anything. Because there is only one type of rods, they cannot distinguish different colors from one another - this is the reason why we cannot see colors in the dark. [3, 4, 6]

The three sets of cones are called red, green and blue cones. Each of them has its sensitivity peak in a different part of the light spectrum, which enables us to distinguish colors (Fig. 1a). The signals from these cones travel to our visual center in the brain in an unusual manner and here is where the perception of color starts. Instead of transporting a separate signal from each type of cones, we combine these signals to form three opponent signals. The first opponent signal indicates the extent to which the green cones have been stimulated opposed to the stimulated red and blue cones, the second signal indicates the balance of blue color versus yellow (which is the sum of green and red cones) and the third carries the information about brightness, i.e. black and white balance. This process of converting signals from the cones to the three opponent signals is schematically represented in Fig. 1b. The mixing of signals from different sets of cones can be understood with the help of additive mixing of colors, as shown in Fig. 2a. If we mix, for instance, red and green light, we obtain yellow light. If we then add blue light to it, we get white. Conversely, if we take the blue part of white light away, we get yellow. [3, 4]
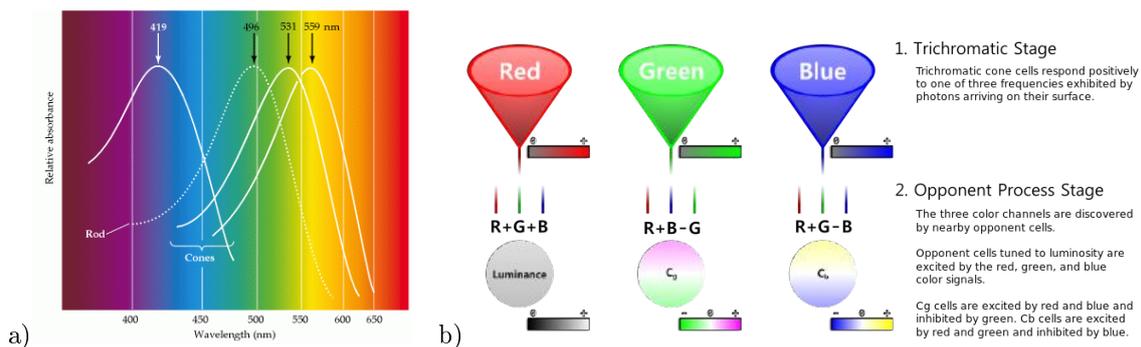

Figure 1: a) The spectral sensitivity of rods (dotted line) and of the three sets of cones (full lines from left to right: blue, green and red). Note that the ordinate axis is not absolute sensitivity, but normalized, so that all peaks have the same absorbance. In reality, rods are approximately 100 times more sensitive than cones. [5] b) A scheme of how the three opponent signals are combined from signals from three sets of cones. [6]

Absorption and emission are two mechanisms which cause coloration. Coloration by absorption is caused by molecules that have broad resonances in the visible part of the spectrum - from 1.6 eV to 3.2 eV. They absorb those frequencies from the incident light and reflect and transmit all others (Fig. 2b). Thus energy of light is partially transformed into thermal energy or the excitations of electrons or atoms. [3, 4] The opposite happens with emission - at nonzero temperatures the electrons, atoms and molecules vibrate and radiate because every charged, accelerating particle radiates. Emission of photons can also occur when an electron, atom or molecule transits from a higher excited state to a state with lower energy. In the range of visible light, these processes are mostly due to electron excitations.
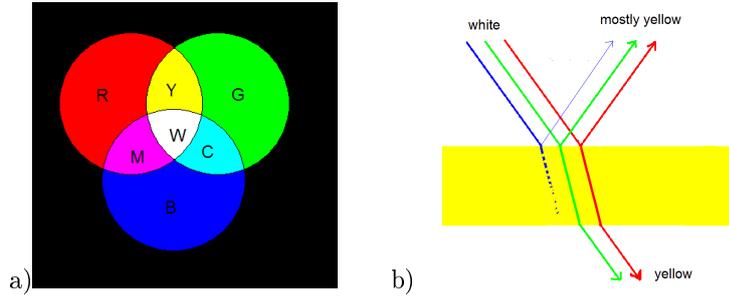
Figure 2: a) The basic principles of additive color mixing - the capital letters represent the colors: R(ed), Y(ellow), G(reen), M(agenta), W(hite), C(yan) and B(lue). b) A substance, which absorbs blue light, appears to be yellow.

3 Structural colors

Structural colors are usually divided into iridescent and non-iridescent. Iridescent colors change strongly with the viewing angle and are typically produced by scattering of light on periodic structures. [7] In this case, the phases of different scattered light waves are correlated to one another, which results in constructive interference of light with distinct wavelengths in distinct directions. An example of these are multiple thin layers and photonic crystals. On the other hand, the colors of random structures are not iridescent, as the phases of scattered waves are not correlated.

3.1 Thin layers

3.1.1 Single layer

We start with the elementary example of structural colors, i.e. interference of a single frequency electromagnetic plane wave $\mathbf{E_i} = \mathbf{E_0} e^{i\omega t - i\mathbf{k_0} \cdot \mathbf{r}}$ which impinges on a thin layer with the angle θ_1. The thickness of the layer is d and its refractive index is n_2. The refractive index of the surrounding media is $n_1 \neq n_2$. θ_2 can be calculated using Snell's law $n_1 sin\theta_1 = n_2 sin\theta_2$, and the amplitude of reflected light is the sum of multiple beams, as can be seen in Fig. 3.
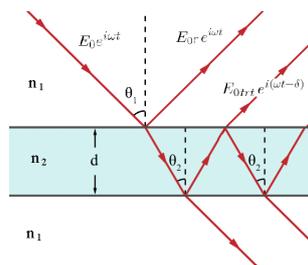

Figure 3: Thin film interference. [8]

Using simple algebra one can calculate the reflectivity of the non-absorbing thin layer, the intensity reflectivity R being defined as the ratio between the intensity of reflected and incident light.

$$R = \frac{2r^2(1 - cos\delta)}{(1 + r^4) - 2r^2 cos\delta},$$

r is the amplitude reflectivity between the layer and the surrounding media and $\delta = k_0\,2\,n_2\,d\,cos\theta_2$ is the phase difference between the reflected waves. Note that it is dependent not only on the characteristics of the thin layer but also on the wavelength of the incident wave $\lambda_0 = \frac{2\pi}{k_0}$ and the gazing angle. This calculation is valid only in an idealized case when the incident light is completely coherent - in reality, the coherence length of sunlight is approximately one wavelength. In this case, two waves do not interfere with each other if their difference in optical paths is larger than a few wavelengths. The joint intensity is then the sum of the intensities of separate waves.

Thin films have a metallic look because the reflected light is directional and their color changes depending on the viewing angle - this is called iridescence.

If the reflectivity of such a system is fairly low, we can consider only the interference between the two strongest wave fronts - the first is reflected off the first interface between the medium and air $E_{0r}e^{i\omega t}$ and the second is transmitted through the medium, reflected on the other side of the thin layer and then transmitted into the air $E_{0trt}e^{i(\omega t - \delta)}$. [4]

The condition for their constructive interference is $\Delta d = \frac{\delta}{k_0} = \delta\frac{\lambda_0}{2\pi} = 2\,n_2\,d\,cos\theta_2 = (m - \frac{1}{2})\lambda$, where m is a whole number and Δd is the so called retardation - the difference in optical paths of the two waves. We can observe that for different colors this condition will be met at different retardations. The explanation of the color sequence, which is often seen on soap bubbles (Fig. 4) arises from this observation. This color sequence is known as Newton's colors.
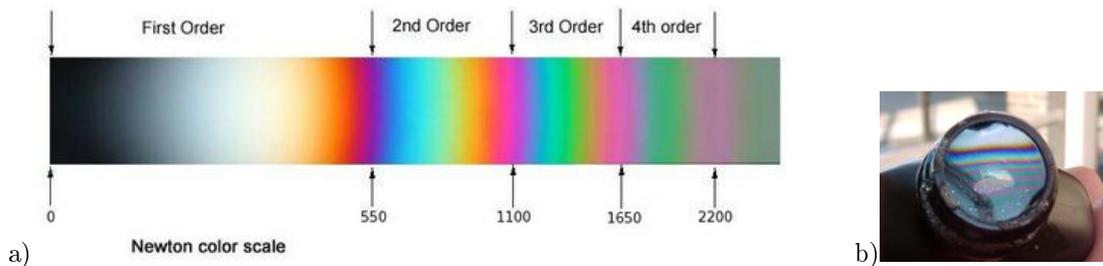

Figure 4: a) Newton's colors. The numbers refer to the retardation Δd in nanometers. [9] b) Newton's colors as seen on a thin soap film. [10]

If we plot the relative intensity and retardation Δd for three distinct wavelengths - red, green and blue, as in Fig. 5, we see that different retardations produce different combinations of red, green and blue light. This results in different colors.

In the case when the thin film is on a surface with a larger refractive index than the film, the condition for constructive interference is modified due to an additional phase shift from the reflection and the order of the colors consequently changes. [3]
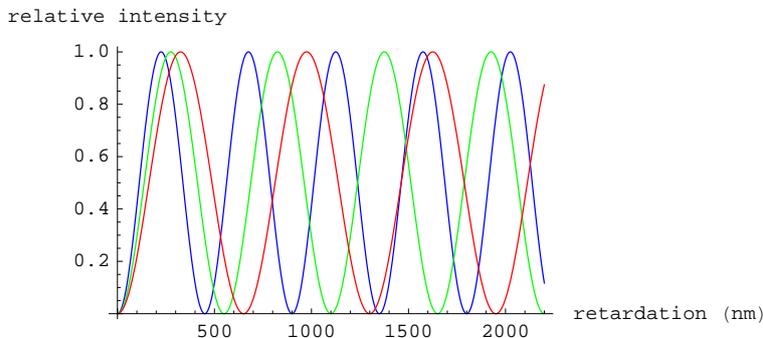

Figure 5: Relative intensity of reflected light for three wavelengths - blue (450 nm), green (550 nm) and red (650 nm), plotted in Mathematica.

3.1.2 Multiple layers

Multilayers are stacks of - in principle - different single layers. In this case, the exact physical formulation of light propagation is much more complex than it was in the previous section. One method for calculating the reflected or transmitted light is by using transfer matrices [2]. For each layer, a matrix is calculated which connects the electrical and magnetic fields on both boundaries for a certain polarization. Then, the matrices for all layers are multiplied and the total reflectivity is calculated.

The other frequently used numerical method is the iterative method [2]. Here, two wave amplitudes for each layer are defined - the waves travel perpendicularly to the boundary of the layers in opposite directions. Their amplitude is described with the reflectance and transmittance of the media, phase gain and the amplitudes of waves in the neighboring layers. From these equations, a recursive formula is derived which connects the reflectivity of a layer with the reflectivity of one of its neighbors. Now, the overall reflectivity can be calculated.

Multilayers are often divided into narrow-band and broadband multilayers. The former usually consist of just a few types of layers and therefore strongly reflect a narrow range of frequencies (Fig. 6). If the lowest layer is dark, it absorbs all the light which has not yet been reflected, thus making the reflected color very vivid. As the base layer becomes lighter, it reflects more and more light of all frequencies, which is then transmitted to the upper layers, resulting in a whiter color of reflected light. Broadband multilayers, on the other hand, reflect light all or almost all wavelengths, regardless of absorption on the base layer, thus having a silver look or, for instance, a gold one, if they reflect all colors but blue and violet. In nature, they usually assist in camouflage in diffusive light. A good example of this are fish scales. [11]

Further examples of colors produced by multiple thin layers are the iridescence of shells, the colors of many birds and insects, etc. [1, 2, 3, 12]
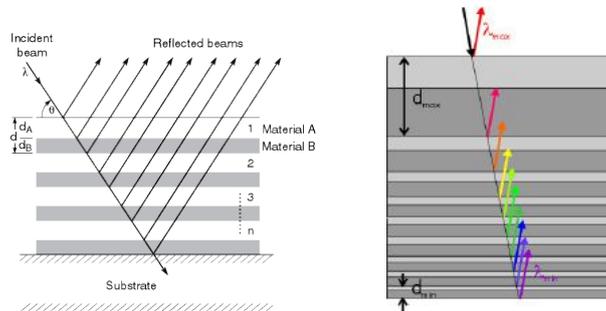

Figure 6: A narrow-band (left) [13] and broadband multilayer (right) [14].

The applications of multilayer films are also plentiful. For example, because it is difficult to reproduce their iridescence, they are often used on banknotes to prevent forgery. Such color-shifting inks are usually made by applying thin layers with appropriate refractive indexes onto a substrate, which is then dissolved away. The remaining multilayer of thickness $\approx 1\,\mu m$ is broken into flakes approximately $10\,\mu m$ in diameter. If these are dispersed in ink or paint, the flakes uniformly fill the media and end up similarly oriented, when the paint is brushed or sprayed onto a surface. With this simple procedure it is possible to produce inks and paints that change their color depending on the gazing angle (Fig. 7). Such paints are becoming quite popular in the industry and are now used in textiles, cosmetics, car paints, plastics and even the coatings of certain machine parts to help spot mechanical damage (Fig. 7c). Also, multilayers are used for antireflective coatings to prevent unwanted reflections off the surface of lenses. [3, 4, 15, 16]

Figure 7: Color shifting ink changes its color depending on the viewing angle. Selected examples include color shifting ink, used (a) against counterfeit [17], (b) for decoration [18], and (c) on machine parts to help spot mechanical damage [19].
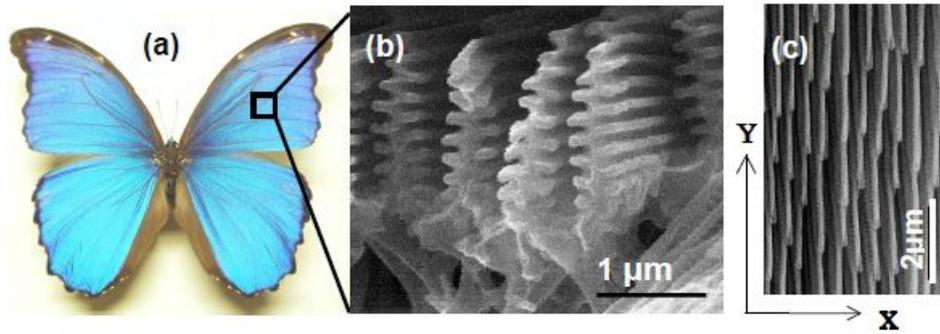

Figure 8: A Morpho butterfly (a) and SEM images of the surface structure of its wings in cross section (b) and from the top (c). [1]

3.2 Photonic crystals

Photonic crystals are materials with a periodic refractive index. [2, 20] This field of research can be described as a mixture of solid state physics and the electromagnetic field - we observe phenomena which arise when an electromagnetic wave and a low-loss photonic crystal interact with each other. The periodicity of the refractive index should be approximately half of the wavelength of the electromagnetic wave in order for it to be diffracted. Photonic crystals are typically divided into 1-dimensional photonic crystals (which are actually a periodic multilayer film), 2-dimensional and 3-dimensional photonic crystals (Fig. 9). [20]
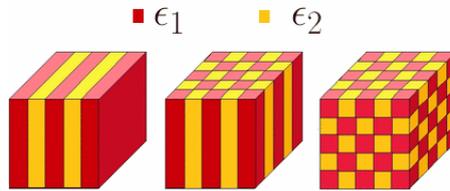

Figure 9: From left to right: 1-dimensional, 2-dimensional and 3-dimensional photonic crystals; the different colors represent materials with different dielectric constants. [21]

Using Maxwell's equations, one can derive the master equation for the propagation of light with a certain frequency ω through media with a periodic dielectric constant $\epsilon(\boldsymbol{r}) = \epsilon(\mathbf{r} + \mathbf{R})$, where $\mathbf{R}$ is a lattice vector.

$$\nabla \times \left(\frac{1}{(\mu(\mathbf{r}))} \nabla \times \mathbf{E}(\mathbf{r}) \right) = \left(\frac{\omega}{c} \right)^2 \epsilon(\mathbf{r}) \mathbf{E}(\mathbf{r}), \tag{1}$$

7

$\mathbf{E}(\mathbf{r})$ is the electric field. In typical photonic crystal materials, $\mu(\mathbf{r})$ is equal to one and ϵ is a scalar. Along with the Maxwell equation $\nabla \cdot (\epsilon(\mathbf{r}) E(r)) = 0$, the master equation determines the electric field. The magnetic field $\mathbf{H}(\mathbf{r})$ can then be obtained using the equation

$$\mathbf{H}(\mathbf{r}) = \frac{-i}{\omega \mu_0 \mu(\mathbf{r})} \nabla \times \mathbf{E}(\mathbf{r}).$$

Photonic crystals can be described by using the Bloch formalism: [22] we define a Bloch state $\mathbf{E_k}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}}\mathbf{u_k}(\mathbf{r})$, $\mathbf{u_k}(\mathbf{r})$ being a periodic function $\mathbf{u_k}(\mathbf{r}) = \mathbf{u_k}(\mathbf{r} + \mathbf{R})$ for all lattice vectors $\mathbf{R}$. We then insert our ansatz into the master equation (1):

$$(i\mathbf{k}\times\nabla) \times (i\mathbf{k}\times\nabla) \times \mathbf{u_k}(\mathbf{r}) = \left(\frac{\omega(\mathbf{k})}{c}\right)^2 \epsilon(\mathbf{r})\, \mathbf{u_k}(\mathbf{r}) \tag{2}$$

Using computational techniques we can calculate the band structure $\omega_n(\mathbf{k})$ and draw a photonic band diagram of the allowed frequencies ω and their relation to the wave vector $\mathbf{k}$. En example is shown in Fig. 10.

For a 1D photonic crystal, the master equation can be analytically solved to a good level. Suppose light is traveling perpendicular to the layers (parallel to the z axis) and it is polarized in x direction. There are two types of layers - the first has $\epsilon = \epsilon_1$ and thickness a, the other $\epsilon = \epsilon_2$ and thickness b (Fig. 10). With the ansatz $\mathbf{E}(\mathbf{r}) = (E(z), 0, 0)$, the master equation is simplified into $-\frac{\partial E(z)}{\partial z} = \left(\frac{\omega}{c}\right)^2 \epsilon(z) E(z)$. We can write the electric field as $\mathbf{E} = \left(Ae^{ikz} + Be^{-ikz}, 0, 0\right)$ for the first type of layer and $\mathbf{E} = \left(Ce^{iKz} + De^{-iKz}, 0, 0\right)$ for the second one. When we apply boundary conditions $E_{x1} = E_{x2}$ and $\nabla \times \mathbf{E}_1 = \nabla \times \mathbf{E}_2$ at $z = a$ and $z = a + b$, we obtain a transcendent equation for the allowed values of K and k.

$$-\frac{(K^2 + k^2)}{2Kk}\sin(Kb)sin(ka) + \cos(Kb)cos(ka) = \cos(ka + kb)$$

Sometimes the left hand side of this equation is greater than 1 or less than -1. In this case we say that the crystal has a band gap - an interval of frequencies in which no real wave vector $\mathbf{k} \in \mathbb{R}$ solves the master equation. This means that light with this frequency cannot propagate through our medium, which is just a different way of saying that the crystal reflects light with frequencies in the photonic band gap.
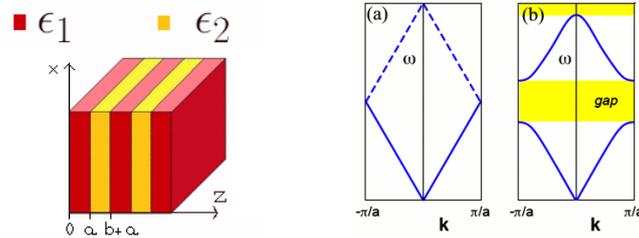

Figure 10: Left: A scheme of a 1-dimensional photonic crystal. Right: a) A photonic band diagram for a uniform medium. [21] b) A scheme of a photonic band diagram of a 1-dimensional photonic crystal. [23]

Using this knowledge, one can design photonic-crystal fibers - photonic crystals with a hollow core, as the ones in Fig. 11. For certain wavelengths, the inner walls appear as if they were a mirror and consequently guide the light along the fiber. What gives these photonic crystal fibers an advantage over "normal" optical fibers is that they have a hollow core, which reduces energy loss. Also, the fibers can be constructed to have desired nonlinear effects, which is promising for their use in optical computers. [20]
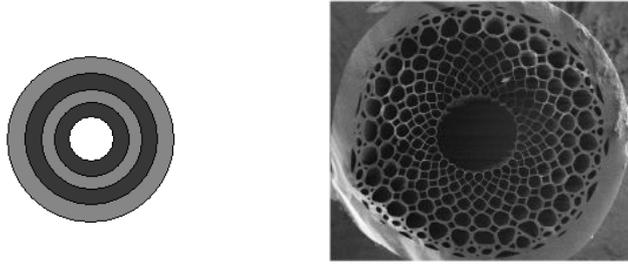
Figure 11: Cross sections of two types of photonic crystal fibers. Light is guided along the hollow core due to the photonic crystal cladding, which acts as a mirror for certain intervals of wavelengths. [24]

Photonic crystals have also been found in nature - the colors of peacock's feathers have been found to be the result of two-dimensional photonic crystals in the barbules of their feathers. The iridescence and bright colors of opals are the consequence of a fcc structure of silica spheres in silica-water. Their phase diagrams have narrow and very directional gaps, therefore they only reflect distinct colors at distinct angles. [1, 2, 20]
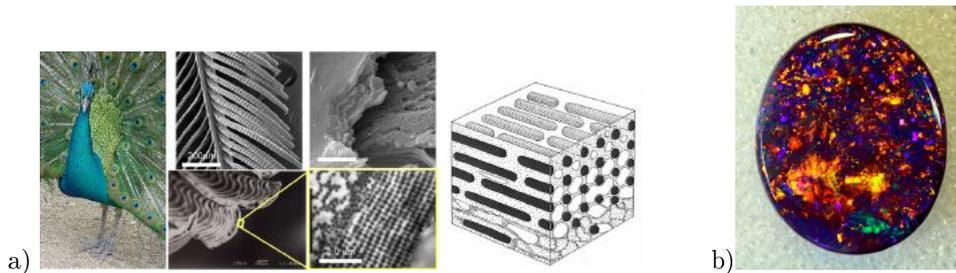

Figure 12: a) from left to right: A peacock, an image of its barbules, their cross section and their interior. [2] b) Natural opals have very bright and iridescent colors. [25]

3.3 Scattering

Scattering changes the trajectory of light due to interaction of light with non-uniformity of the media. It is the elementary mechanism, by which flow of light is tuned. Indeed, diffraction, reflection and transmission are macroscopic phenomena which have their roots in scattering. In this seminar we focus only on elastic scattering on microscopic particles, thus conditioning the scattered wave to have the same wavelength as the incident. [4]

There are two limit regimes of scattering: scattering on particles, which are much smaller than the wavelength of incident light, is called Rayleigh scattering. In this case, the ratio between scattered light and incident light is different for various frequencies. The other limit is when particles are larger than the wavelength. This is often called Mie scattering and results in a white color of material.

3.3.1 Rayleigh scattering

Imagine the interaction between a polarized electromagnetic wave with the wave vector $\mathbf{k}$, wavelength λ and frequency ω, $\mathbf{E_i} = \mathbf{E_0}e^{i\omega t - i\mathbf{k}\cdot\mathbf{r}}$, and an atom. The light wave induces a time-varying (Hertz) dipole on the atom $\mathbf{p} = \alpha\mathbf{E_i} = e\mathbf{d}$, which then radiates accordingly, as seen in Fig. 13. The intensity of the radiation is

$$I_{radiated} = I_{incident}\left(\frac{\sin\phi}{4\pi R}\left(\frac{\omega}{c}\right)^2\alpha\right)^2.$$
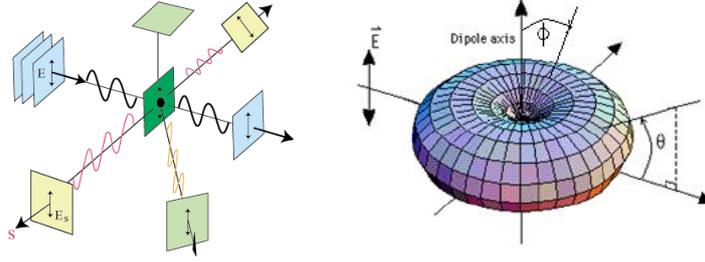
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Figure 13: The polarization (left) and intensity (right) of dipole radiation. [26] [27]

For a non-polarized electromagnetic wave and a fairly small particle ($d \lesssim \lambda/15$), each of the atoms in the particle radiates as a dipole and the phase differences between them are practically negligible, because the distance between any two atoms in the particle is much smaller than the wavelength of incident light. They interfere constructively and the particle therefore acts as one big dipole. The dipole axis changes randomly in time because the incident wave is not polarized. The time-averaged intensity of scattered light is now of the form

$$I_{scattered} = I_{incident} \frac{1 + \cos^2 \theta}{2R^2} \left(\frac{\omega}{c}\right)^4 \left(\frac{n^2 - 1}{n^2 + 2}\right)^2 \left(\frac{d}{2}\right)^6 , \tag{3}$$

θ being the angle between the direction of the incident wave and the viewing point, R is the distance between the dipole and the viewing point and d is the diameter of the particle. The terms $\left(\frac{n^2-1}{n^2+2}\right)^2 \left(\frac{d}{2}\right)^6$ come from the Clausius - Mossotti relation $\mathcal{N}\alpha = 3\frac{n^2-1}{n^2+2}$, where $\mathcal{N}$ denotes the number of atoms in the volume of the particle with a refractive index n. The angular dependency of scattered light can be observed in Fig. 15. The final equation for the outgoing wave is a sum of the scattered wave and the remaining of the incident wave. [2, 4, 12]

If we look at a tenuous gas with no absorption in the visible, at an angle, perpendicular to a white incident beam, we see linearly polarized violet light with some blue. The color is due to the fact that intensity of scattered light is proportionate to ω^4, so violet and blue scattered light have a higher intensity than orange and red. When we look directly towards the incident light, we see the medium as if it were red / orange, which is white light minus the scattered violet and blue. The exact color depends on the relative amount of scattered light.
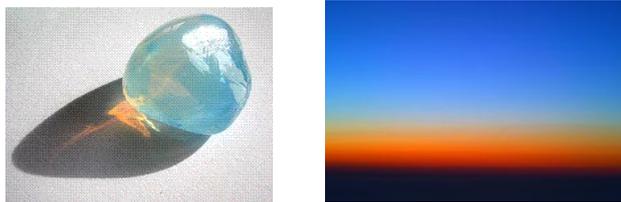

Figure 14: The effect of scattering on colloidal particles, known as Tyndall scattering, is similar to Rayleigh scattering. Tyndall scattering in opalescent glass (left) and Rayleigh scattering in the sky (right). [28] [29]

Rayleigh scattering nicely explains the blue color of the sky. According to the previous paragraph, one might expect the sky to be violet, but it is not. The reason lies in the the absorption of UV and violet light by ozone in the atmosphere and Sun's emission spectrum, which contains more blue than violet. Therefore, although violet light is more effectively scattered than blue, the latter prevails. With knowledge of Rayleigh scattering the yellow-white color of midday Sun and the red sunsets are also easily explained. At noon the light from the Sun travels through a much

thinner layer of air than at sunset, so less blue and UV light are scattered and more of them reach the viewer. This results in a yellow hue, opposed to a red hue at sunsets. [3]

The greater the concentration of microscopic particles, the greater is the likelihood of scattering. This is why redder sunsets are seen after major volcanic eruptions or where the air is more polluted with particles, smaller than 100 nm (so called ultra-fine particles).

3.3.2 Mie scattering

Consider now the case when the particles are somewhat larger than the wavelengths of incoming light, which is called the Mie scattering regime. Now, the phases of different dipoles do not necessarily interfere constructively. This is the reason that, as the size of particles grows, less and less light is scattered backwards and to the side. In other words, light reflects, refracts and diffracts from the particles. This can be seen in Fig. 15. [4]
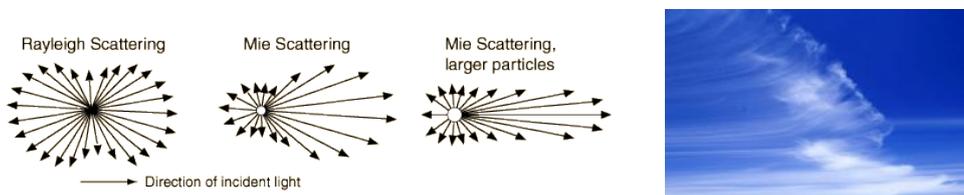

Figure 15: Left: The angular dependency of scattered light intensity for Rayleigh and Mie scattering. [30] Right: Mie scattering on water droplets causes the white color of clouds, whereas Rayleigh scattering on gas molecules results in a blue sky. [31]

For spherical particles $\lambda < d$, the scattered light is only weakly dependent on λ. Therefore, the scattered light contains all wavelengths and appears white. This explains the white color of fog, mists, clouds, snow, salt, sugar, beaten egg white, paper and many more. All consist of transparent particles in a transparent medium. The white color comes from multiple reflections off the boundary between two materials with a different refractive index. [3, 4]

4 Conclusion

In this seminar, we showed the basic structures and physical mechanisms that are responsible for the emergence of structural colors. The structures include single layers, multilayers and photonic crystals, whereas for mechanisms we focused on Rayleigh and Mie scattering. We explained that random structures produce non iridescent colors via scattering, whereas periodic structures result in iridescent colors. In reality, most so called periodic structures have defects of some sort, which can alter the appearance of colors.

Structural colors surround us in our daily life. We can find them in nature - for instance in feathers, the blue color of the sky and butterfly wings - and as well as in man-made objects. They are used in security to prevent forgery, they help show signs of physical ware, guide light when optical fibers cannot, some are used for camouflage, while others for purely aesthetic reasons. The applications of structural colors to industrial products are today to an important extent a result of advances in assembling and controlling nanostructures. Further advances in the field may result in an even wider use of structural colors.

Nowadays, the research of structural coloration materials has moved from simple to more complicated, quasi-ordered structures, metamaterials and structures where more than one mechanism produces the final color. The field of photonic crystals, as one of the fields focusing also on structural colors, is quickly expanding and it promises to play an important role in optical computers, which might some day replace today's electrical current and silicon chip technology.

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