Antisolar windows

This part of the project focuses on the technical analysis of antisolar windows and glass panels for radiative cooling. Antisolar windows, also called solar control windows, reflect a significant portion of the infrared (IR) light, which is responsible for most of the heat. This block aims to provide you with a deep understanding of the way materials can be used in thin films in order to have specific optical properties, and thereby go beyond their intrinsic properties.

Indeed, optical properties emerge from the way light interacts with a system made of a set of materials in a given configuration, giving rise to interference effects that can be used to reinforce or decrease light transmission and reflection at specific wavelengths. Hence, as for electrons in solids, forbidden gaps can exist for light, and dispersion relations can be tuned for photons in materials, similar in concept to electronic bands in solids. This is the field of photonic crystals, which we will only consider here in one dimension corresponding to multilayered materials.

Overview

You will have to acquire new notions and use them to compute the performance of different types of windows. Therefore, you will:

1. learn the link between the optical properties of solids and their electronic and atomic structure;
2. discover the optical properties of the specific materials you will work with in the project;
3. investigate how light is reflected and transmitted by thin (multilayered) films, and how to compute reflectivity, transmittivity and absorptivity. From this, analyze the performance of a specific antisolar window and propose an optimal design;
4. perform lab experiments to characterize antisolar windows, and discover ellipsometry and infrared spectroscopy;
5. learn how heat is radiated by solids, and more specifically by thin coatings. Based on this, investigate whether glass panels capable to radiatively cool a device while being transparent in the visible range are feasible.

You should report on your findings in the wiki. Note that there are a lot to do so you should share the work between the different members of the group. Use your complementary strengths to maximize the efficiency of your group work. But be nevertheless aware that, for the open-book test of this part of the project, everybody is supposed to understand properly the origin of optical properties of solids, the main phenomena which appear in multilayered films, and the basic aspects of thermal radiation.

Resources

You have access to a series of podcasts, reference documents, excel spreadsheets and websites to help you in the project. The podcasts were made for another project such that you may have to adapt what is said to your specific case. Reference books are provided so that you can browse through them when needed to find the useful information. Review articles summarize a topic in an accessible way. Websites can be found by you, but we mention some which are particularly useful. Additionally, we will provide answers to your questions during the planned mentoring sessions of the project. **However, it is your duty to prompt us and ask questions.** Hence, we will help you find your way in the project, but we will not teach anything unless you ask.

Among these resources, there is:

1. `"Radiative heating and cooling with spectrally selective surfaces"` by C. G. Granqvist, *in* Appl. Opt. 20, 2606 (1981). This review article, albeit quite old, presents the main ideas behind antisolar windows in its section IV, and behind radiative cooling in its section III. A more recent review paper by the same author, which might actually be less informative, is `"Solar energy materials for`

thermal applications: A primer" by C. G. Granqvist & G. A. Niklasson, *in* Sol. Energy Mater. Sol. Cells 180, 213-226 (2018).

2. "Optical Properties of Solids" by Mark Fox. This book is an absolute reference for... optical properties of solids. You will certainly dig into this one.

3. "Ellipsometry and Polarized Light" by R. M. A. Azzam and N. M. Bashara, a very good albeit rather mathematical book on ellipsometry. Rest assured, you will not have to read the complete book, but some sections will prove handy to compute the reflection of light by multilayers, and to interpret ellipsometry experiments.

4. We prepared an excel spreadsheet implementing the computation of reflection and transmission of light through a simple substrate/film/ambient system, *i.e.*, implementing the equations of Azzam and Bashara's book. The spreadsheet allows you to check that the results produced by your own codes are correct, thereby allowing you to debug them, but it will not be sufficient for the project. Hence, you better write your own codes! The spreadsheet is read-only; you may want to download it to use it – also note that we used a French-localized version of Excel when writing it, with a decimal period (not a comma).

5. An excerpt of a podcast explaining how to compute the reflection and transmission of light by a multilayered system, which essentially explains in a condensed way section 4.6 of Azzam and Bashara. Beware, the podcast considers the case of a multilayer in vacuum or air; in your case, the bottom layer is not vacuum, but glass – adapt what you'll see in the podcast!

6. "Spectroscopic Ellipsometry and Reflectometry. A User's Guide" by Harland G. Tompkins and William A. McGahan, a practical book on spectroscopic ellipsometry which you may find useful to help you interpret your experimental data. Don't count on it for a precise mathematical treatment of ellipsometry – but experience shows that it is quite helpful for unexperienced ellipsometry users. Or ask your tutor!

7. The first and the second volumes of the handbook "Optical Constants of Solids" by Edward D. Palik, which contain $n - j\kappa$ for a series of materials, in tabular format.

8. On the website refractiveindex.info, you will have access to $n - j\kappa$ tabulated data for many materials – easier to use than to type Palik's data yourself!

9. "Development of Radiative Cooling and its Integration with Buildings: a Comprehensive Review" by J. Chen & L. Lu *in* Solar Energy 212, 125 (2020), a review article on radiative cooling in buildings – the English is unfortunately poor, but the content is better.

10. "Passive radiative cooling below ambient air temperature under direct sunlight" by A. P. Raman, M. A. Anoma, L. Zhu, E. Rephaeli & S. Fan *in* Nature 515, 540 (2014), an article which convincingly demonstrated for the first time the application of the concept of passive radiative cooling;

11. "Thermal Radiation Heat Transfer" by John R. Howell, M. Pinar Mengüç & Robert Siegel. That is a monster of a book, and many parts of it are not useful for you, but it will be handy when you will work on radiative cooling.

12. An excerpt of a podcast explaining how to compute the thermal emission of a body, which essentially explains in a condensed way what you could read in the book of J. R. Howell *et al.* Beware, the podcast considers the case of a sail radiating energy by its two large surfaces, whereas your window should radiate only *via* its external surface; but since you will not have to compute the emitted energy but simply the emission spectrum, this is not important in your context.

13. An excel document with the solar spectrum (at the average latitude of the USA), at sea level, and for the sun at zenith; one sheet of the document also contains the transmittance spectrum of the atmosphere.

14. For programming, you are on your own. But we tried `the copilot.microsoft.com generative IA` and found that it could provide good propositions of Python programs to compute optical properties – provided we entered the complete very descriptive prompts of what we wanted to get, and provided we corrected a few bugs in what was proposed. That might be a proper starting point to spend less time on coding, and more time on thinking...

Your mission

At the end of this period, your wiki should have added sections on your work in this block, in addition to an introduction to solar thermal management in buildings and your LCA analysis (a section is *a set* of pages, not one single page!). Check the starting and closing dates of this block in the `calendar` of the project — and respect deadlines!

The work itself will be a journey through increasingly complex optical systems, split by us in tasks with deadlines; there will be regular tutoring sessions in which we will be present to answer your questions, and explain some concepts if needed to the class – but again, we insist that there will be no formal presentation unless you specifically ask for it.

Task 1 – Introduce the main concepts used when computing the optical properties of antisolar windows

The optical properties of windows depend on the complex index of refraction of the materials of the window, $\tilde{n}(\omega) = n(\omega) - j\kappa(\omega)$ [1].
Compute the reflectivity of an infinitely-thick slab of glass (you can find `here a table of` $n - j\kappa$ `versus wavelength of a typical glass`, taken from Palik), and of an infinitely-thick slab of the metal assigned to your group in `the Table of materials` (panel 'Antisolar glass', column 'metal film') in the wavelength range from 200 nm to 20 μm, for normal incidence, incidence at 35° and incidence at 70° – **for non-polarized light**[2]. If you have a mixture of metals, use Bruggeman's law to compute the average indices of refraction. Plot these data in graphs together with the index of refraction of your materials in the same range, and explain the main features of this graph with respect to the electronic and vibrational properties of the materials. For this, the book of Fox will be helpful. Use a logarithmic axis for the wavelengths, since they vary over two orders of magnitude.

Instead of using the experimental refractive index for your materials, you could compute it using first-principles calculations. To understand the theory behind, you may need to go back to your LMAPR1491 and LMAPR1492 classes. The steps to perform such calculations are described here. Compare the experimental and ab initio-computed inde of refraction of your metal, and comment the differences you observe.

In your wiki, introduce the concepts of antisolar windows (the articles of Granqvist are probably useful here), and start a section on the reflection of light in which your results will be reported.

Task 2 – Thin metallic films on glass as antisolar windows?

A possible way to block most of the solar spectrum except visible light (300–700 nm) is simply to evaporate a thin metal film over the glass (and place this fragile thin film inside the double-glass panel to protect it from scratching).

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[1] Note the minus sign for the imaginary part. This is a common definition used in literature, related to the way the time component of the waves is written. Here, we write a forward-traveling wave as $\exp(j(\omega t - \vec{k}(\omega)\cdot\vec{r}))$; then the index of refraction is $n - j\kappa$ so that the wave is attenuated as it moves forward. Indeed, suppose the field wave travels in the x-direction and can be written as $\vec{E} = \vec{E_0}\exp(j(\omega t - \tilde{k}(\omega)x))$ in which $\tilde{k}(\omega)$ is the (complex) wavenumber. Since $\tilde{k}(\omega) = \omega/\tilde{v}(\omega)$ where $\tilde{v}(\omega) = c/\tilde{n}(\omega)$ is the (complex) speed of light in the considered medium and $\tilde{n}(\omega) = n(\omega) - j\kappa(\omega)$ is the complex index of refraction of the medium, $\vec{E} = \vec{E_0}\exp[j(\omega t - \tilde{n}(\omega/c)x)]$. Then, $\vec{E} = \vec{E_0}\exp[j(\omega t - n(\omega/c)x)]\exp(j^2\kappa(\omega)x) = \vec{E_0}\exp[-\kappa(\omega)x]\exp[j(\omega t - n(\omega/c)x)]$, which shows that the wave is attenuated exponentially when progressing in the medium. If we had taken $\tilde{n} = n + j\kappa$, the wave would have been exponentially amplified when passing through the medium. An alternative convention would be to write a forward-traveling wave as $\exp[j(\vec{k}\vec{r} - \omega t)]$; then the index of refraction should be taken as $\tilde{n}(\omega) = n(\omega) + j\kappa(\omega)$ to attenuate a forward-progressing wave. If you are interested by conventions, have a look to `this article by R. H. Muller`.

[2] If you wonder how to compute the reflectivity for a non-polarized light, check `this document`.

1. Write a program to compute the transmission and reflection spectra of such a window at normal incidence for a selected value of metal film thickness, and plot the reflectivity (or reflectance) R, the transmissivity (or transmittance) T and the absorbance $A = 1-R-T$ of the air/film/glass interface for different thicknesses between 0 and 100 nm.[3] Then compute the percentage of visible and non-visible light of the solar spectrum that will be reflected, transmitted or absorbed by the film at the external surface of the window, depending on the thickness of the metal film – ignore the thickness of the glass and consider non-polarized light, and plot these percentages versus film thickness. From this, select an optimal thickness for the metal film. The metal we propose your group to use is again in `the Table of materials` (panel "Antisolar glass", column "metal film"). Section 4.3 of Azzam & Bashara, or sections C3–C5 of Tompkins & McGahan provide the theoretical background to compute the reflection and transmission of thin films on a substrate. You can use the spreadsheet we made to check your computations.

2. Obviously, normal incidence is not the only possibility. Perform the same computation for a vertical window oriented towards the Sun in Brussels, on June 21 noon. If you wonder how the Earth is positioned versus the Sun at that date, `have a look here`. Then compare the results obtained by each group or try yourself different metals. Which metal would be best suited? In your comparison, take into account cost and environmental incidence.

3. Report on your findings in the wiki, and indicate the limits of this simple technology.

4. Meanwhile, you should have received samples of glass coated by a thin layer of an unknown metal; each group will have a sample with a different metal layer thickness. Take appointments with Delphine Magnin and Sabine Bebelman and measure your samples by spectroscopic ellipsometry and infrared spectroscopy (in reflection), respectively. You may ask your tutor to explain in more details what is ellipsometry. For ellipsometry, use as wide a wavelength range as possible, and measure at two angles of incidence, 35 and 70°; for IR, measure in reflection at normal incidence over the widest wavelength range; actually, why not try in transmission as well? Then analyze your data:

   (a) for ellipsometry, trace the two (Ψ, Δ) trajectories obtained for each incidence angle, using wavelength as parameter. Find a way to obtain the thickness of your thin film and the values of $(n - i\kappa)$ versus wavelength. Can you identify the metal?

   (b) for infrared reflection, use the knowledge obtained by ellipsometry to compare the measured values with the values you can predict.

   (c) report on your observations and deductions in your wiki – making links to what you wrote already!

Task 3 – Can interferences be used to improve antisolar properties?

If we want to have efficient antisolar windows, we clearly need something more sophisticated than a simple metal film on glass. By adding more layers, we can actually tune reflection and transmission using interferences between the many reflected and transmitted beams in the structure. You can totally extinguish a wavelength or reinforce another one using this concept. This is extensively used by glass producers, one of the simplest applications being antireflection coatings (as used on the glass panels protecting solar cells). In this task, we propose you to investigate such multilayered systems.

1. In `the Table of materials` (panel 'Antisolar glass', column 'trilayer'), you will find a trilayer structure assigned to your group. Write a program to compute the optical transmission and reflection by multilayered structures for any angle of incidence, and use it to explore how it is possible to transmit as much light as possible in the visible range while reflecting the rest of the solar spectrum. For this part, the podcast and section 4.6 of the book of Azzam and Bashara might prove useful. Find optimal values for the thickness of each layer for the case of normal incidence, and compute the performance of your optimal window compared to bare glass or the simple metal film on glass for normal incidence.

   What is the physical reason a trilayer behaves better than a single layer?

[3]The reflectance is the ratio of reflected power to incident power; the transmittance is the ratio of transmitted power to incident power; and the absorbance is the ratio of absorbed power to incident power. To see how to compute them from the Fresnel coefficients of reflection and transmission of the field, read `this short note which defines intensity and power`....

2. You are now in the position to evaluate how much solar power is reflected by your windows compared to bare glass, and thereby estimate the electricity you will spare for air-conditioning. This number should be used to complement your life-cycle analysis of glass windows – in another block of the project.

3. You will have received samples from AGC, covered by metallic oxides – use ellipsometry and infrared spectroscopy to characterize their performance. You have access to `the public technical sheet` describing glass windows incorporating these samples. Is it possible to reverse-engineer these samples and find what they are made of?

4. Then enter a largely unexplored technology, in which the trilayer system would be replaced by a multilayer alternating thin layers of a dielectric material and thin layers of an aerogel (for which you can approximate the index of refraction to the one of air). The nature of the dielectric we propose is as always in `the Table of materials` (panel 'Antisolar glass', column 'multilayers - dielectric'). Here, you are on your own: you may want to explore the effect of the number of layers (we recommend 10 bilayers), of their thickness, or the possible interest of a distribution of thickness on the performance of the coating. For instance, you may draw 2D maps of the performance (color scale) versus thickness of aerogel (x axis) and thickness of dielectric (y axis), for a given number of layers; work at normal incidence but consider also the possible side effects of such multilayers when changing the angle of incidence. Here, thinking how to display your results is crucial.

Report on your work in your wiki, explain your observations, and discuss the merits and drawbacks of each technology.

Task 4 – Exploring radiative cooling

In this part of the project, we propose you to investigate whether it would be possible to coat glass panels of solar cells with coatings that would favor the transmission of the visible photons but would contribute to cool the panels by radiatively emitting heat in the infrared transparency window of the atmosphere. This heat arises from the photons that are not captured to generate electricity and thus heat the panels. Since the efficiency of solar cells decreases at higher temperature, cooling the cells is a good idea. The radiation emitted in the atmospheric transparency window is sent to space and dissipated away from earth. Your colleagues working on the half-project are actually working on 'cool roof paints', some of which also exhibit radiative cooling which decreases the demand on air-conditioning. The materials that are used for this effect typically should emit strongly in the atmosphere transparency window, hence have strong absorption bands for these wavelengths.

Section III of Granqvist (1981), Table 2 of Chen & Lu, or the Nature paper of A. P. Raman *et al.* propose materials and stacks for radiative cooling. You may select freely one and investigate whether it is possible to construct a stack that would provide transparency in the visible range, reflection of the rest of the sun energy, and radiative cooling in the infrared transparency window of the atmosphere. A podcast explains how radiative emission can be computed based on the same equations we already used before, and of course there is the book of Howell, Menguc & Siegel in case you would want to dig deeper. The transmittance spectrum of the atmosphere is also available.

In your wiki, explain the origin of the absorption bands in the 10 μm wavelength range, the structure you imagined and why you selected this specific structure, and your conclusions regarding the feasibility of this idea. Chapter 10 of Fox provides information on IR absorption bands and their link to the phonon dispersion bands of materials. To visualize the associated modes of atomic vibration, you can connect to the `materials project website`, search for your material, and observe atomic vibration modes. Note that not all modes can be used to dissipate heat – you may want to check why in Fox and explain.