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Technology and Materials for Passive Manipulation of the Solar Spectrum in Greenhouses

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Greenhouse horticulture grows increasingly important due to its ability to provide a controlled microclimate which is optimizable for highly efficient crop growth and resource use, although it may come at a significant energy and investment cost. One of the most crucial inputs in any greenhouse is sunlight, giving free energy and light for greenhouse crop growth. However, it is enormously variable, both geographically and seasonally. This review discusses materials and technologies usable in greenhouse cover and screen materials which can passively manipulate the incident sunlight to transmit a light spectrum that is ideal for crop growth, thereby improving the yield, and for greenhouse microclimate management, thereby reducing the energy usage of greenhouses. The current status of spectrum-manipulating technology in greenhouses, developments over the last few years, some potential innovations adaptable from diverse fields to greenhouse horticulture, and the associated challenges, are discussed.

1. Introduction

As the impacts of climate change become more apparent, issues are arising such as unpredictable and drastic weather fluctuations, temperature rise, and impacted freshwater availability. These factors affect agricultural output, and could potentially trigger a food security crisis for the rising global population. In this scenario, greenhouse horticulture is becoming increasingly important due to its ability to provide a stable and controlled microclimate for plant growth, which results in high crop productivity and a high potential for water saving, though it comes at a steep energy and investment cost. Despite a lack of accurate numbers on the total greenhouse area in the world (well in excess of one million hectares), there is agreement on an estimated 6% growth in greenhouse area per year worldwide.[1]

One of the most important inputs in a greenhouse is light, required for crop photosynthesis. Even though an abundance of sunlight is available on earth, free of cost, it is not optimally distributed across space or in time—several regions across the world receive too high or too low solar radiation, which further varies significantly through the year. If greenhouse growers are to make the best use of naturally available sun energy and yet want to maintain a consistent output yield, the sun radiation entering the greenhouse requires some manipulations, especially since the energy carried by sun radiation also determines the microclimate inside the greenhouse. Only a small fraction of greenhouses worldwide are equipped with active climate control installations (heating, cooling, etc.); most are "passive" greenhouses depending on the incoming sun radiation and energy transmitted through, and thus influenced by, greenhouse covers and screens.[2] Innovation in the area of such passive technologies that can, with lower material, installation, and maintenance costs, boost the yield and quality of crops, reduce the energy consumption of greenhouse production, and are designed keeping safety and sustainability in mind, are broadly in alignment with UN Sustainable Development Goals 2 (Zero hunger), 3 (Good health and well-being), 7 (affordable and clean energy), 10 (Sustainable cities and communities), 12 (Responsible consumption and production), and 13 (Climate action).

Some spectrum manipulation techniques applied to greenhouse covers have been in place for a long time—such as seasonally applied shading paints and nets in regions receiving high sunlight, and anti-reflection coatings to maximize sunlight transmission in regions receiving low sunlight. While these do aid greenhouse productivity and crop protection, there is significant scope for improvement. Additionally, several of these methods are not the most sustainable options in terms of their maintenance, replacement, and disposal. In the past few years, with increasing focus on sustainable development goals and the energy transition, new materials, technologies and developments have emerged that might enable further fine-tuning of the incident sunlight in greenhouses, in terms of both intensity and spectrum.[3]

Greenhouse technology stands to benefit from recent innovations in closely related fields such as photovoltaics (PV) and energy-efficient buildings, such as luminescent solar concentrators (LSCs),[4–6] semi-transparent organic semiconductors,[7–9] smart windows,[10–13] and self-cleaning windows.[14,15] Additionally, several adaptations could also be derived from developments in technologies like photon upconversion[16–19]

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or quantum dots (QDs),[20–24] which are commonly applied in diverse fields like bioimaging, biosensing, optical devices, or display technology. So far, to a large extent, the translation of these materials and technology to greenhouses either lacks rigorous experimental trials or large-scale commercial implementation. Moreover, such a translation requires the recognition and resolution of challenges specific to the context of greenhouses. Identifying promising light manipulating technologies and assessing their potential to be adapted in greenhouse covers is the primary objective of this review. To this end, we have compiled some recent advances in spectrum manipulation which are potentially adaptable to greenhouse cover materials or screens, that might benefit plant growth and development as well as greenhouse microclimate, consequently resulting in higher yields and/or energy savings. Keeping the objective of energy saving in mind, the technologies described here are passive, that is, do not require any additional energy input for spectrum manipulation. Active technologies,[3,12,13,25] which can switch their optical properties in response to stimuli generated via an energy input, for instance, electrical voltage pulses, are not considered here.

The review begins with a brief discussion on the solar spectrum incident in a greenhouse and the effects of the different spectral components on plant growth and greenhouse microclimate. A short overview of conventional spectrum manipulation practices is followed by recent technological developments adaptable to greenhouses: In each case, a brief description is followed by the current status, possibilities, and the main challenges in the context of greenhouses. Scientific publications—especially those where plant or greenhouse trials have been carried out—are highlighted. Studies reporting spectrum manipulation in alternate applications such as smart windows, PV technology, and LSCs, which can be translated to greenhouses, is also included. The terminology has been kept general to be accessible to a wide range of readers in the field of greenhouse technology—the main intention is to present a compilation of relevant promising technology and the scope for innovation, rather than providing a deep physical explanation. For in-depth scientific and technical details regarding the various technologies, the reader is pointed to the cited literature.

At the outset, it is important to mention that even though we only review simpler, passive means of spectrum manipulations, several of these materials and technologies are still quite far from attaining the size and production costs that greenhouse applications would require, and thus their widespread commercial implementation might not be possible in the short term. However, our goal is to highlight here some highly promising directions for innovation in this field, which, following extensive and systematic research, might eventually be able to match the steeply growing demand for sustainable, fossil-fuel free, energy-efficient systems.

2. The Solar Spectrum inside a Greenhouse

The solar spectrum received on the earth surface, as shown in **Figure 1**a, is composed of ≈5% ultraviolet (UV) radiation (250–400 nm), 45% of photosynthetically active radiation (PAR) (400–700 nm, which is very nearly the band visible to our eyes), and ≈50% of near infrared (NIR) radiation (700–2500 nm). Depending on the location and cloud cover, the relative proportions of these components can vary to some extent. These components affect the growth and development of plants inside the greenhouse, either by triggering a variety of plant photoreceptor pigments or by affecting the greenhouse microclimate through the energy they contain. The main effect of the UV component is on plant growth and development. On one hand, UV radiation can be beneficial, when leading to production of secondary metabolites such as flavonoids, phenolics, carotenoids, and ascorbic acid,[26,27] potentially leading to better colors of leaves, flowers, and fruits, better taste, and higher resilience against pest and diseases. On the other hand, it can cause DNA damage and inhibit photosynthesis, reducing growth and inducing compactness.[28] The NIR component mainly influences the greenhouse microclimate by its heating effect: the incoming NIR is significantly absorbed by the interior of the greenhouse, but only partly by plants—≈40% of the NIR energy is reflected by leaves.[29] The PAR component contributes significantly to adding more energy to the greenhouse, but also has a major contribution to plant growth and development through photosynthesis and photomorphogenetic effects. The majority of crops are sun-loving crops (e.g., tomato), for which the greenhouse productivity is known to increase almost linearly with the intensity of PAR radiation received,[30] making maximized PAR transmission one of the most crucial considerations in greenhouse design for most crops. For shade-loving crops (e.g., diverse flower crops and lettuce), photosynthesis is saturated at much lower PAR intensities[31,32] and there is a higher scope of observing photomorphogenetic effects by replacing wavelength-neutral shading strategies with spectrum-modifying shading.[31]

The entire band of PAR radiation does not contribute equally to photosynthesis. The pigments responsible for photosynthesis have absorption peaks in the blue and red part of the spectrum—for chlorophyll a, these lie around 430 and 662 nm, and for chlorophyll b, around 453 and 642 nm (Figure 1b). These wavelengths are most efficiently utilized by plants for the photochemical process of photosynthesis. However, on the crop level, absorption and utilization of a broad range of photons of ≈400–700 nm contribute to photosynthesis.[33] Apart from this, different bands of light activate secondary photosynthetic pigments such as carotenoids, and non-photosynthetic receptor pigments like phytochromes, cryptochromes, and phototropins already at low intensities, which can impact all aspects of plant growth, development, and metabolism including seed germination, plant shape and morphology, flavor production, and nutritional quality (see Figure 1c). Most of the activating wavelengths lie in the blue part of the spectrum (400–500 nm) or in the red (600–700 nm) and far-red (700–800 nm) part of the spectrum. Green light (500–600 nm) is absorbed to a somewhat lesser extent by these pigments, leading to the green appearance of leaves. Though its role in photosynthesis is not as critical as the red or blue wavelength bands, its ability to penetrate through a plant canopy much more than the blue or red bands is important to excite photosynthesis in the lower lying leaves in the canopy.[34]

Recent developments in light-emitting diode (LED) technology have led to a more precise identification of the roles of different

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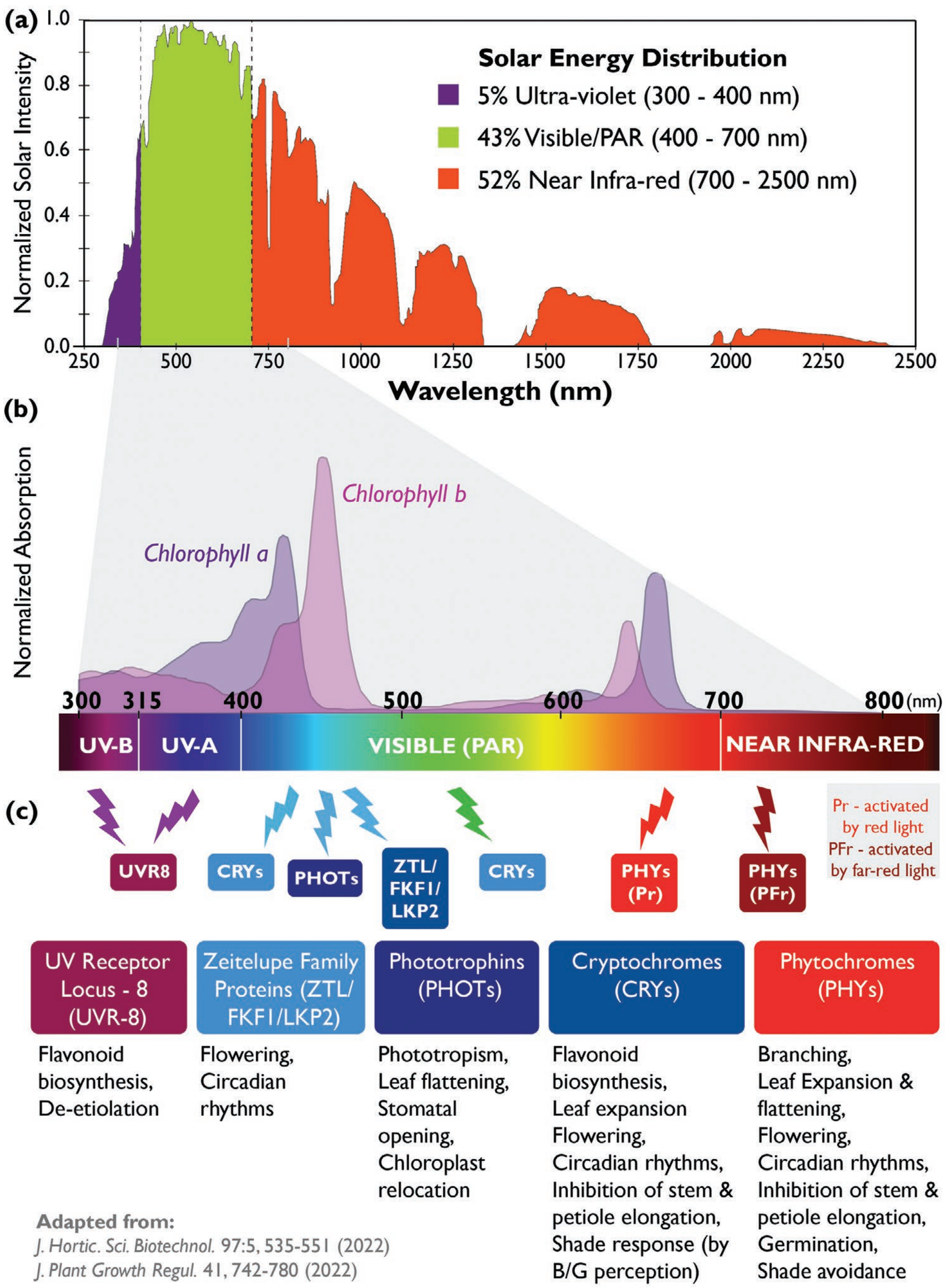

Figure 1. a) Typical spectrum of solar radiation incident on the earth surface (after atmospheric absorption). The solar spectrum is divided into the components—UV, PAR, and NIR – shaded in the colors purple, green, and red, respectively. b) Normalized absorption spectra of chlorophyll a and b pigments responsible for photosynthesis. c) Wavelength specificity of the major plant photoreceptors shown along with the plant growth/development/morphological parameters affected by the respective pigments. Adapted with permission under terms of the CC-BY license.[39] Copyright 2022, the Authors, published by Springer Nature. Adapted with permission.[37] Copyright 2022, Taylor and Francis Group.

wavelength bands in plant growth and development processes by enabling selective, finely tunable control of the light spectrum composition. The effect of the LED spectrum on various parameters such as plant biomass and morphogenesis, photosynthesis, yield, flowering, secondary metabolites, and nutritional quality has been extensively studied at both genetic and physiological levels, and compiled into several detailed reviews.[35–38] An overview of spectral effects on plant growth and development is shown in Figure 1c (adapted from refs. [37,39]).

The insights derived from controlled LED illumination provide useful starting points toward tailoring spectrum manipulation for specific desired outcomes. However, it remains to be verified how representative these results are of plant processes in real greenhouse conditions (i.e., a full spectrum and albeit

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modified). The processes activated by each wavelength band of the spectrum would act in concert inside the green house, and their interplay could give rise to emergent effects that might be different from the selectively excited processes under LED illumination. Moreover, LED-based experiments also reveal differences in wavelength-dependent physiological responses in different plant species, highlighting the need to characterize the effects of the same spectrum-manipulation technology for a variety of crop species, while opening up possibilities of crop-specific design of these technologies.

3. Solar Spectrum Manipulation Technologies

Some typical outcomes expected from spectrum manipulation in greenhouses are: a boost in the yield, selection for desired photomorphogenetic or qualitative properties in crops (color/compactness/secondary metabolite content), energy-saving in greenhouse operation, and creation of a suitable microclimate in the greenhouse. The latter is especially different for different locations:[40] in tropical areas with high solar irradiance, the excess heat needs to be removed, whereas in temperate regions, especially during cold seasons with lower solar irradiance, heat retention is desired.

Knowing the role of different spectral components of solar radiation in the greenhouse, we can take two approaches to manipulate the spectrum: (i) selectively transmitting some spectral components through the greenhouse while blocking the rest, either by reflection, scattering, or absorption—this can be done using scattering nanoparticles, 1D photonic crystals (1DPCs), or plasmonic nanoparticles; (ii) conversion or shifting of the less desirable wavelengths into wavelengths that are more "useful" for photosynthesis, plant growth, development, morphogenic effects, or greenhouse microclimate regulation. Such conversion can be done by utilizing the phenomena of luminescent downconversion or upconversion.

3.1. Conventional Spectrum Manipulation Practices

The most commonly used materials in greenhouse coverings so far operate by scattering the incident light to reduce its transmission. One example is the seasonal whitewash used in high-light-intensity regions, consisting of a mixture of calcium oxide and calcium carbonate nanoparticles which reduce sunlight transmission in all wavebands. While such coatings have been reported to show positive effects on regulation of greenhouse temperature, reduction of the cooling load, decrease in transpiration, and improvement of the water-use efficiency,[41] the loss of transmission in the PAR band limits the greenhouse yield. Moreover, spatial homogeneity of application is a challenge for such coatings. Many high-sunlight-intensity regions also use shading nets for light scattering. A detailed review[26] on the use of shading nets in horticulture reports positive effects on crop yield, and by using colored nets, even on photomorphogenetic effects, across a wide variety of crops. Though the relative transmission of the different spectral wavebands can be modulated using colored nets, the overall PAR light transmission reduces by at least 50%.[26] Moreover, while these nets lead to both shading and spectral selectivity, the contribution of these two factors to the overall effects of such netting have not been separately analyzed, preventing a complete understanding of the exact effects of spectral manipulation by these nets.

To combat PAR loss, there have been developments on introducing partial spectral selectivity, especially focused on the rejection of the NIR waveband as its heating effect in the greenhouse is particularly undesirable in summers with high sunlight intensity—this has been attempted using films with NIR-absorbing or NIR-reflecting pigments, and NIR-filtering spray-on whitewash coatings.[42–44] A comparative study of several such materials showed that NIR-reflection is more effective for thermal management than NIR-absorption, due to the heating, and eventual emission, of NIR-absorbing covers. Moreover, it was suggested that the effect of NIR reflection of 800–1100 nm wavelengths was more significant than of 1100–2500 nm wavelengths (as the 800–1100 nm wavelength band has higher energy and is also present with a larger intensity in the solar spectrum compared to the 1100–2500 nm band), and that efficient NIR-filtering still had significant scope for improvement.[45]

Light rejection in a narrow wave-band has been studied for UV wavelengths in greenhouse coverings[46] and also in applications like windows and food packaging.[47,48] Many of these studies employ nanoparticles of TiO_2 and ZnO to selectively absorb UV radiation due to their electronic band gap.[47,49,50] However, PAR transmission is not as critical a factor for windows and food-packaging as it is for greenhouses, thus the materials from the cited studies cannot be directly adapted in greenhouses without optimizing for maximum PAR transmission. Blocking sunlight in well-defined wavelength windows without compromising on the transmission of other wavebands is a crucial requirement for spectral-selective greenhouse covers. Such selectivity can be achieved by using materials like 1DPCs, discussed in Section 3.2.

3.2. One-Dimensional Photonic Crystals

Photonic crystals are artificially engineered materials designed in a way such that their dielectric function, and thus refractive index, varies periodically across the structure of the material along one or more dimensions, affecting light propagation through these materials. 1DPCs have such periodic variation only in 1D: essentially, these are multilayers with alternating layers of high and low refractive index, respectively (for instance alternating metal and dielectric layers). This periodic modulation of the refractive index results in successive back-reflections of incident light at each interface. The structural parameters of a given 1DPC define a range of wavelengths called the "photonic band gap" (PBG)—for wavelengths within this range, the back-reflections are in-phase and interfere constructively, so that the majority of the incident light is reflected or "rejected" from transmitting through the photonic crystal. For wavelengths outside the "PBG," the out-of-phase back-reflections interfere destructively—the majority of the light is thus "allowed" to transmit through the crystal.[51–54] The PBG can be tuned by either changing the refractive index "contrast"

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between the high- and low-refractive index layers (by changing the material composition of the layers), or by changing the periodicity at which the refractive index is modulated, (by varying the relative or absolute layer thickness, number of layers, or by using a more complex repeating pattern).

Multilayer materials have been extensively researched for application in heat-shielding windows in buildings,[10,11,14,48,55–57] and the same principles can be adapted for thermal management in greenhouses as well. Some of the earliest multilayer systems for NIR blocking involved alternating metal and dielectric layers,[58] which can make for high costs of depositions on large surfaces like greenhouse covers. Recently, there has been considerable research on polymer-based 1DPCs that can be lighter, more flexible and easier and cheaper to manufacture.[48,59,60] Despite the promise of better mechanical properties, comparative ease of sourcing of raw materials, and reduced fabrication costs, the biggest drawback of polymer-based photonic crystals is a much lower contrast attainable between the refractive indices of adjacent layers compared to inorganic systems—this affects both the bandgap width, as well as the effectiveness of spectral blocking, which is far from inorganic 1DPCs and requires improvement. An important consideration while adapting 1DPCs to greenhouses is that the PBG, which is determined by the interference of the back reflections at the multilayer interfaces, varies with the angle of incidence of light.[61] Thus, the precise position and width of the PBG might not be maintained for the differently oriented greenhouse faces, the varying direction of the incident light across the day and year, and for diffuse sunlight. This can be accounted for by designing wide band gap PCs,[48] particularly in the broad NIR waveband. The promise of 1DPCs lies in their "customizable" optical properties, for instance, through computational optimization.[56,62,63] In particular, a recent publication[64] capitalizes on this to precisely engineer a multilayered filter composed of the materials Al_2O_3, ZnS, and Ag, that can transmit a spectrum optimized for maximum yield of lettuce (*Lactuca sativa*) through LED experiments, and simultaneously blocks NIR transmission almost entirely. Such plant-specific multilayer filters, possibly deployable as screens, could result in high individual yields for different crops grown under the same generic natural or artificial light sources.

A big challenge for these materials is combining large-scale fabrication with precise layer thicknesses, often on the nanometer scale. Though industrial scale fabrication for such structures has been developing in the last few years,[60] the complexity, and thus cost, increase for multilayers with increasing customization of their optical properties. Further, a larger number of multilayer repeats are required for such tailoring, which then has the adverse effect of lowering the overall transmission through the multilayer. Moreover, these materials operate by blocking light in the first place, and hence have limited potential for greenhouse covers in low-light conditions.

A different system providing spectrally selective transmission of light, with the additional possibility of spectrally selective absorption, is plasmonic systems, which can, instead of reflecting away the undesirable wavelengths, make use of them by converting them to heat.

3.3. Plasmonics

The field of plasmonics is based on the phenomenon of "plasmon resonances:" collective resonant oscillations of free electrons triggered by shining electromagnetic radiation on the plasmonic system. In the context of this review, we discuss "localized" plasmon resonances—electron oscillations confined to the surface of metallic nanoparticles, rather than "propagating" surface plasmons—waves of electron plasma propagating on a continuous interface between a metal and a dielectric.[65–68] Typical systems showing such localized plasmon resonances are gold or silver nanoparticles, which have a large density of nearly free electrons that oscillate with the electric field of incident light.[65] Exciting these oscillations resonantly results in dramatically enhanced electric fields or "hot spots" in the near-field of the nanoparticles, along with highly amplified absorption or scattering behavior at the resonance wavelength. The shape, size, and material of the nanoparticles, and the properties of the surrounding medium determine the resonance wavelength—the tunability of these parameters leads to tunable spectral selectivity in these systems. The collective oscillations excited in the plasmonic nanoparticles decay nonradiatively as heat, which makes plasmons a suitable system for efficient light-to-heat conversion.[69,70]

Both, the spectrally varying optical properties and light-to-heat conversion are applicable to greenhouse technology. For instance plasmon-based visibly transparent, UV and NIR-blocking filters have been computationally designed[71] for PV cells by combining plasmons of different dimensions.[71] Such nanostructures with well-defined shape and order, could prove challenging and expensive to fabricate precisely on a large-scale fabrication, on top of the already high costs of working with metals like Au and Ag. However, recent work on IR-blocking glass embedded with disordered plasmonic nanostructures composed of inexpensive materials like Cu, TiN, or Al, presents promising possibilities for relatively inexpensive passive cooling of buildings, and potentially also greenhouses.[72]

Plasmonic light-to-heat conversion, meanwhile, can find application in low-energy heating in greenhouses located in cold climates: combined with the large absorption and thermal conversion of the incident light (facilitated by the enhanced absorption cross section of the nanoparticles at plasmon resonance), the wavelength selectivity of plasmons can be exploited to tune the resonance wavelength to coincide with relatively less useful wavelengths of the solar spectrum, which can then be selectively converted to heat. A few studies have computationally and even experimentally designed plasmon-based nanoheaters embedded in low thermal conductivity materials such as aerogels to retain the plasmon-generated heat, leading to high increases in local temperature under low incident light intensity.[73,74] Simulations made for NIR absorbing plasmon-based films in a southern-Spain summer climate indicated a temperature rise of 3 °C due to absorption[75]—undesirable for a southern-Spain greenhouse in summer, but useful for passive heating in colder climates. The effective heating at greenhouse length-scales in sunlight and temperature conditions for colder climates needs to be experimentally quantified, though. plasmonic-heating could also be used to drive defogging[76] of

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the covering materials, thus restoring the transmission lost due to condensation on the cover.[77] Due to their strong near field generation, plasmons are often used in conjunction with other processes to enhance the optical/scattering cross sections and, in fact, have been used in combination with other spectrum manipulation technology, as described in Section 3.5.

The biggest disadvantage of plasmon-based technology is the cost, especially when using noble metals like Au or Ag. A growing body of research on plasmonic properties of aluminum, an inexpensive as well as earth-abundant metal, will improve the prospect of commercial viability of plasmonic systems, but also their sustainability.[78,79] The high heating effects at plasmon resonance could cause melting of the nanoparticles at high light intensities, altering their shape and size, and consequently the spectral properties and performance of these systems. Good thermal management is then important in the design of these systems to avoid damages to the nanoparticles or the medium that they are embedded in. While the light-to-heat conversion resulting from the plasmon near-field enhancement can facilitate energy-saving in greenhouses, there is no effective "enhancement" of PAR intensity to boost photosynthesis, and thus, yield. Increasing the photosynthetic photon flux density is achievable using spectral conversion driven by luminescent process such as photon down- and up-conversion.

3.4. Luminescence

"Luminescence" is the phenomenon of emission of light. The emission can be triggered by a variety of stimuli like electrical energy, mechanical stress, chemical reactions, or absorption of photons. The latter phenomenon is known as "photoluminescence." We look at the two kinds of photoluminescence that can be applied to greenhouse technology: downconversion and upconversion. Simple schematics depicting a generalized concept of the energy transfer for both processes are shown in **Figure 2**a,d. The actual energy transfer process can be much more complex, involving many more components and steps (Figure 2b,e). Technical details of the mechanism, composition, and design of suitable luminescent materials can be found in several references[17,80–82] which cover a wide variety of applications, ranging from fluorescence microscopy to PV technology to artificial photosynthesis. In the following sections, we briefly discuss both phenomena specifically in the context of greenhouse covering and screen materials, and mention applications where plant or greenhouse trials have already been carried out and some promising results from other fields that might be interesting to adapt to greenhouses.

3.4.1. Downconversion

Luminescent downconverting materials, commonly known as fluorescent materials, absorb photons of a certain energy, and after some internal conversion processes, emit lower energy photons (Note that for the sake of simplicity, we use the term "downconversion" for the situation where the emitted photons have lower energy than the absorbed photon—this can happen via two different mechanisms, termed as downconversion and down-shifting, the details of which can be found in references such as [81]). A simple schematic of this process is shown in Figure 2a. Figure 2b shows the energy diagram associated with a more realistic material—in this case, a dye based on the organic compound coumarin.[83,84] Unlike the conceptual energy diagram, a realistic molecule has several vibrational energy levels associated with each energy level, which leads to a broadening of the absorption and emission spectra around the peak wavelengths of absorption (λ_{abs}) and emission (λ_{em}), as seen in Figure 2c. As blue and red are the most important wavelengths for photosynthesis, the ideal spectral downconversions for greenhouses are UV (λ_{abs}) → blue/red (λ_{em}), or green (λ_{abs}) → red (λ_{em}).

Two important parameters determining the performance of fluorescent materials, or fluorophores, for greenhouse applications are (i) fluorescence yield, that is, the efficiency of spectral conversion, and (ii) losses resulting from self-absorption, also known as Stokes losses. The latter results from overlap between absorption and emission spectrum (see Figure 2c). The extent of overlap is determined by the Stokes' shift, that is, the energy difference between the peak absorption and peak emission: the higher the spectral overlap, the more the absorption of the emitted intensity by the fluorophore itself.

The most common materials showing downconversion are organic dyes, QDs, and lanthanide ions.[85] Organic dye-based fluorophores include derivatives of organic compounds such as rhodamine, perylene, and coumarin, among several others.[4] An important organic dye commonly used in both academic research and commercial products for greenhouses is Lumogen F Red 305 (LR305), specially designed for LSCs—fluorescent panels that channel the emitted light to PV components for electricity generation from sunlight. LR305 has a high fluorescence yield and good photostability, but a narrow absorption range and high spectral overlap.[4,86]

The discovery of QDs, crystalline nanoparticles made of semiconductor materials, improved the prospects of tuning the absorption and emission spectra of fluorophores significantly. QDs typically have narrow emission spectra which can be tuned by varying the size, composition, and through doping or surface coatings.[20] This tunability can be used to engineer QDs with very low self-absorption losses, increasing the overall fluorescence efficiency.[87] Other advantages of QDs over organic dyes include greater fluorescence yields and better stability against degradation.[88–90] Quantum dots typically have a broad absorption spectrum—this does not pose any significant disadvantages for LSCs which can utilize the entirety of the broad wavelength range for electricity generation. However, in greenhouses, excessive absorption in the blue waveband for downconversion to the red waveband can significantly affect the morphology and secondary metabolite production in the crop. Some general disadvantages include higher fabrication costs, difficulty in integration with a host matrix, and potential toxicity arising from heavy metals such as lead (Pb) and cadmium (Cd).[91]

Lanthanide ions like neodymium (Nd^{3+}), ytterbium (Yb^{3+}), and europium (Eu^{3+}) can also be used for luminescent downconversion, either as a combination of two ions, or combined with organic molecules, but the main disadvantages are low absorption coefficients and expensive prices.[85,91]

The idea of using fluorescent films to optimize plant growth in greenhouses is not recent—plastic films with fluorescent

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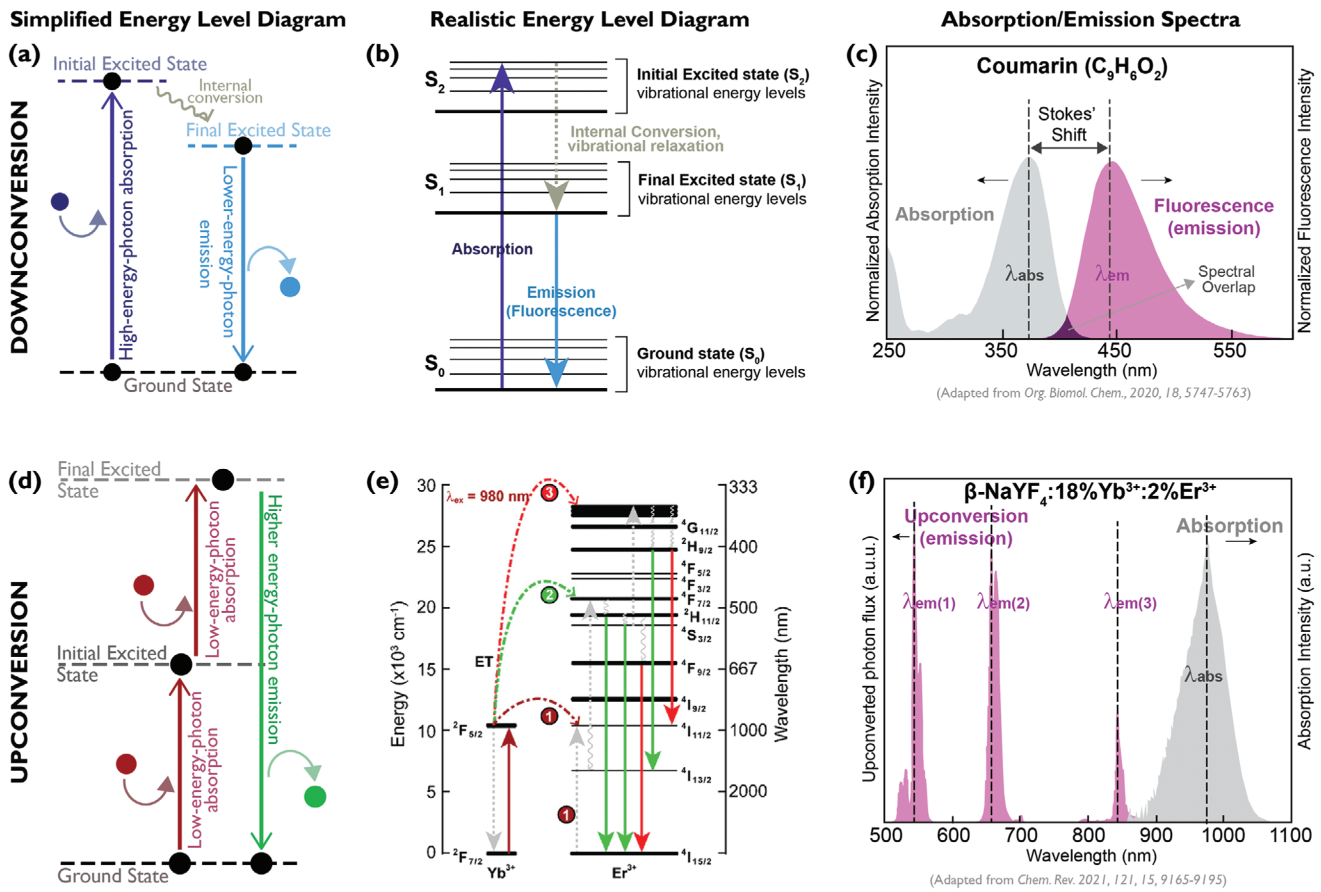

Figure 2. Luminescent downconversion and upconversion processes: a,d) simplified conceptual illustrations. Realistic energy transfer diagrams corresponding to materials are shown in (b) for a coumarin ($C_9H_6O_2$)-based organic dye and (e) for a rare-earth-based upconversion material, β-$NaYF_4$:18% Yb^{3+}/2% Er^{3+} having Yb^{3+} ions as sensitizer, Er^{3+} ions as the emitter and $NaYF_4$ as the substrate. The colored arrows indicate energy transfer processes occurring between different energy levels in the materials. c,f) Fluorescent, upconverted emission spectra plotted (pink curves) alongside the absorption spectra (grey curves) for c) the coumarin-based organic dye and f) the β-$NaYF_4$:18% Yb^{3+}/2% Er^{3+} upconversion material. The absorption wavelengths (λ_{abs}), emission wavelengths (λ_{em}), Stokes' shift, and spectral overlap are indicated in the figure. c) Adapted with permission.[83] Copyright 2020, Royal Society of Chemistry. e,f) Adapted with permission.[82] Copyright 2021, American Chemical Society.

pigments were tested as greenhouse covers for a variety of crops including tomatoes and strawberries since as early as 1990, with generally positive results.[92–98] However, possibly due to recent developments of efficient and more stable fluorophores with tunable spectra, coupled with the growing knowledge of light spectral effects on crops, fluorescent materials have once again become an active area of research in greenhouse technology, with several academic publications just within the last 5 years,[99–106] some reporting as high as 20% increase in crop fresh weight. Commercial manufacturing involving integration of fluorophores in greenhouse cover or screen materials has also seen considerable progress recently[107–111] with claims of positive effects on leaf and stem size, fruit size, and even numbers for a variety of crops such as lettuce, strawberries, tomatoes, and cannabis.

Some of the key challenges for fluorescent films (and ongoing research to solve them) are summarized below:

i) Low fluorescent yield and self-absorption losses of fluorophores (solutions: increasing efficiency of luminophores,[99,100] lowering Stokes losses[87,112])
ii) Trapping of emitted light within the film due to total internal reflection (solution: higher light-extraction in one direction by introducing structural deformations on one of the film surfaces[113,114])
iii) Poor luminescence lifetime of the fluorophore (solution: enhancement of photostability and luminophore lifetime[99,100])
iv) Potentially toxicity, for example, with lead or cadmium-based QDs (solution: cadmium-free QDs made with carbon, silicon, or other heavy-metal free compositions[85] and sustainable fluorophores made of earth-abundant or organic materials.[115])

The factors (i) and (ii) considerably reduce the total transmitted photon flux density through fluorescent films. However, some studies with well-quantified photon flux density with and without fluorescent films report 8.7% increase in dry weight, 11% increase in fresh weight, and 13% increase in leaf area of lettuce despite reduction of 12% in the daily light integral (DLI) measured for photons in the PAR range[103] and around 19–22% increase in the fresh and dry weight of lettuce despite a reduction of 20% in the DLI[106] under these films. This could

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possibly arise from an increase in the red/far-red ratio, which has been said to enhance developmental activity and regulate stem elongation rates and branching.[93] Understanding the exact mechanism of yield increase via fluorescent films, however, requires careful accounting of two additional effects that occur intrinsically in these films: shading, or effective reduction of the transmitted photon flux, and diffusion, arising due to isotropic photon emission by the fluorophores. The effect of shading can be determined by measuring the light intensity, either in form of the photon flux density or the DLI, inside and outside the films, and also by conducting the experiments, wherever possible, under both low and high light intensity conditions, as done in this study.[106] To separate the intrinsic diffusion of the fluorescent films from the diffusion of the substrate itself, control studies should use an identically diffusive substrate without the fluorophore (for instance, in the study in ref. [113]). Both shading and diffusion can significantly impact the yield, especially in regions receiving high light intensity[43,116,117] and for shade-tolerant crops.[118] Unfortunately, careful control for these parameters seems to be missing in several reports of plant/greenhouse trials with spectral conversion films, similar to the case of photoselective nets. The difference in spectral requirements of different plant species also necessitates extensive characterization of the effects of such films across a variety of crops and variants.[38]

For natural sunlight, another challenge is the difficulty of attaining significant photon downconversion to the blue band, since wavelength bands with more energy than blue constitute a very small part of the solar spectrum. Photons in the red/far-red band are much easier to obtain by downconverting either out of the UV, blue, or green wavebands. While the red band is important for plant extension and biomass increase, even small enhancements in the photon density in the blue and UV band of the spectrum might show interesting effects on photomorphogenetic properties (color and compactness, important for ornamental plants), and on secondary metabolites controlling flavor and nutritive content of the crops. This limitation, fundamentally imposed on spectral downconversion of solar radiation, can be overcome by a phenomenon called luminescent upconversion, described in Section 3.4.2.

3.4.2. Luminescent Upconversion

Luminescent upconversion involves the absorption of several photons with low energy and the consequent emission of a photon of higher energy. Upconversion proceeds via a more complex mechanism than downconversion. A simple picture of upconversion involves the successive absorption of several low energy photons by the upconverting system, the resulting excitation of its electrons to a higher energy level, and relaxation of electrons from this energy level via the emission of a high energy photon (Figure 2d). However, the actual energy transfer diagram can be quite complex (see Figure 2e/ref. [82]) and involves several components to facilitate the process. For instance, commonly studied rare-earth ion-based upconversion systems based involve: an activator or emitter ion (with several available metastable energy states in a ladder-like configuration, allowing successive excitations of electrons in multiple energy transfer steps); a sensitizer ion (which undergoes excitation by the incident low-energy photons and resonantly transfers energy to the activator ion to facilitate multi-step excitations); and a substrate (a host material selected to minimize non-radiative energy losses such as by heating, and maximize the radiative emission of high energy photons).[16] The rare-earth ions erbium (Er^{3+}), thulium (Tm^{3+}), and holmium (Ho^{3+}), are commonly used as activators due to their ladder-like arranged energy levels. Ytterbium (Yb^{3+}) is commonly used as a sensitizer due to its large absorption cross section, and for its ease of energy transfer with the above-mentioned activator ions. A commonly used substrate material is $NaYF_4$, and in general chloride, bromide, and fluoride-based materials can be used as host matrices. Organic systems can also show upconversion, driven by a slightly different mechanism driving the upconversion, based on sensitizer and annihilator/emitter molecules. Material choices for upconversion and the associated energy transfer processes are described in detail in the reviews.[119,120] In general, the desired upconversion in greenhouses would be NIR (λ_{abs}) → red/blue/UV (λ_{em}), or green (λ_{abs}) → blue/UV (λ_{em}). Both these conversions have been reported under low power artificial illumination.[120,121] Upconverting solar radiation in greenhouses still seems far from commercial realization. In applications like bioimaging, medical therapy, and lasers,[120] upconversion generally involves illumination with coherent, intense, highly focused laser sources. A critical analysis of upconversion for PV technology[82] found that the upconversion efficiency needs a few orders of magnitude improvement for it to be practically viable for PV. However, for greenhouses, even small increases in the PAR photon flux density by upconversion might yield noticeable results, and will benefit from more proof-of-concept experiments. While a small upconversion to UV photons might helpful in color/flavor/nutrient boosting, NIR-to-visible photon upconversion offers the possibility to combine thermal management and photosynthetic photon flux enhancement. Some pilot studies of the effect of upconverting systems combined with photosynthetic systems are promising—a 12% increase in photosynthetic rate has been observed in *Arabidopsis thaliana* under a film containing both downconverting luminophores as well as NIR → PAR upconverting luminophores under natural light supplemented with UV;[122] NIR → PAR upconverting films have been reported to increase the leaf number (12.5%), leaf area (33%), and chlorophyll content in tomato plants under artificial lighting[123]—these studies provide a starting point for materials whose properties and efficiency could be optimized further, and tested for a variety of crops under natural solar illumination. Efforts to improve the upconversion performance of materials under lower light intensities equivalent to the incident solar radiation intensity on earth, currently involve experimenting with material parameters, nanostructuring, use of photonic structures such as dielectric microlenses to concentrate the incident light, and using plasmonic structures to get local field enhancements.[82,119] These results provide some much needed first steps in the direction of making upconversion a viable technology for greenhouses under natural sunlight. With respect to sustainability and safety, organic material-based upconverting systems might be preferable to rare-earth-based systems, and are also reported to have larger absorption cross sections, efficiencies, and tunability.[16] The range of emission

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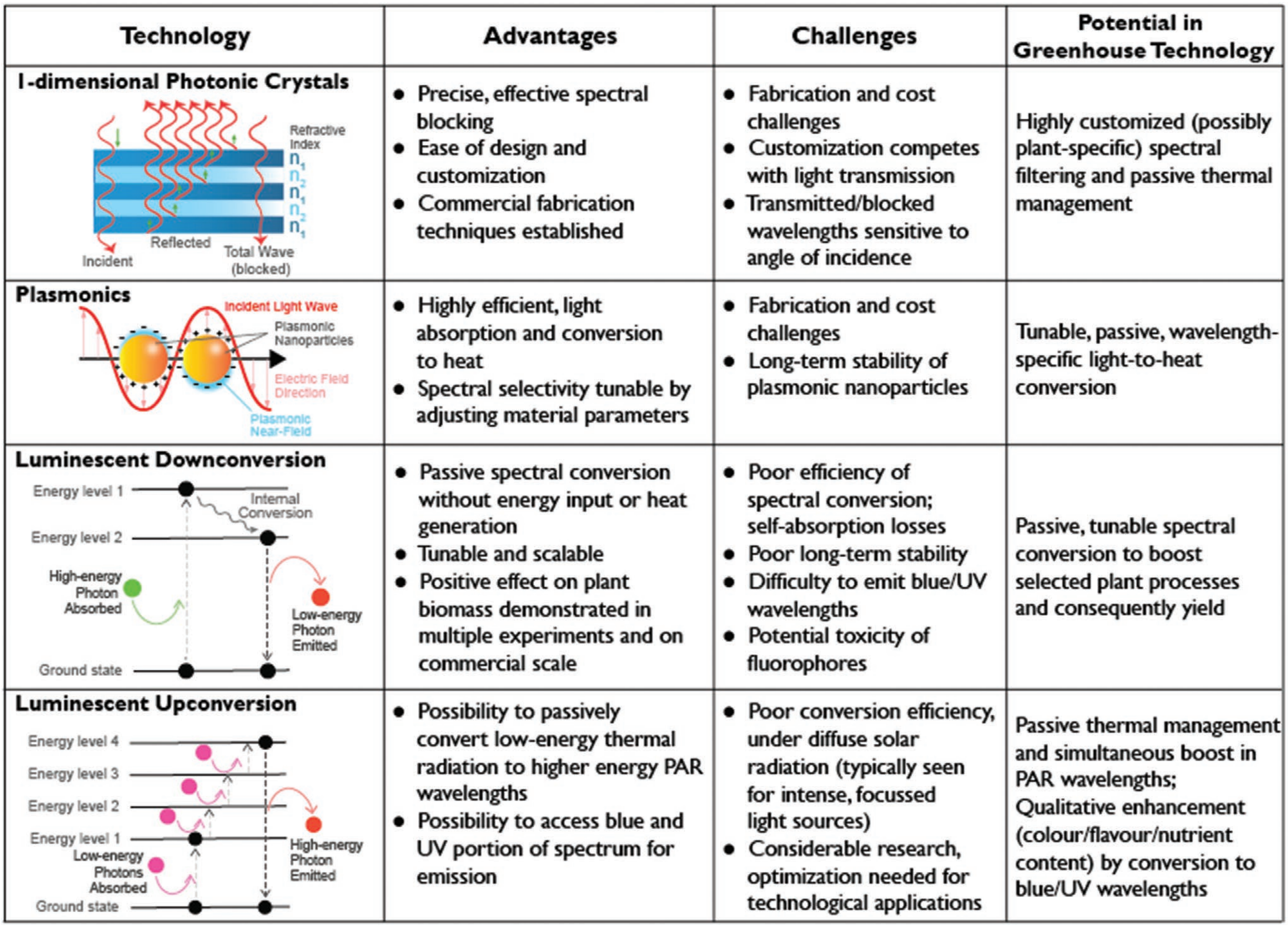
wavelengths observed for organic upconverting systems was 380–630 nm, whereas for rare-earth-based upconversion systems it was 290–800 nm, both of which are promising for PAR enhancement in the greenhouse. However, the stability of organic upconverting systems needs considerable improvement. Moreover, for the purposes of thermal management in greenhouses, materials based on rare-earth elements such as those containing Er- or Yb-sensitizers work better due to the excitation wavelengths of 980 nm for Yb^{3+} and 1500 nm for Er^{3+}[82] whereas despite using different combinations of sensitizers and emitters, excitation wavelengths have been obtained only in the range of 420–780 nm (as of 2015)[16] and optimization and research is needed to have the possibility of NIR-excitation,[16,124] particularly in the 800–1100 nm wavelength range, as discussed in Section 2.

3.5. Possible Combinations of Technologies

The previous sections discuss a variety of spectrum manipulation technologies, each with its own set of advantages and drawbacks, as summarized in **Figure 3**. With the goal of working around the limitations of each technology to transmit the most optimal spectrum into a greenhouse, several recent reports have featured combinations of different technologies to design complex covering materials. For instance, 1DPCs have been combined with fluorescent films to decrease the backscattering in one direction,[61] and also to combine NIR blocking with visible (PAR) spectrum manipulation.[104] The latter study[104] also combines two different downconverting organic dyes to broaden the absorption spectrum in the visible range to effectively increase the fluorescent emitted photon flux. Plasmonic materials have been combined with both up- and down-converting films to enhance the local fields around the luminescent ions/molecules to increase emission.[120,125] Combining CdZnSe QDs with Au nanoparticles in polymeric films was even reported to lead to an enhanced luminescence lifetime,[101] and warrants further exploration. Additionally, beyond the scope of this review, the growing field of agrivoltaics attempts to combine spectral manipulation for photosynthesis enhancement with generation of electricity.[126]



Technology	Advantages	Challenges	Potential in Greenhouse Technology
1-dimensional Photonic Crystals	• Precise, effective spectral blocking • Ease of design and customization • Commercial fabrication techniques established	• Fabrication and cost challenges • Customization competes with light transmission • Transmitted/blocked wavelengths sensitive to angle of incidence	Highly customized (possibly plant-specific) spectral filtering and passive thermal management
Plasmonics	• Highly efficient, light absorption and conversion to heat • Spectral selectivity tunable by adjusting material parameters	• Fabrication and cost challenges • Long-term stability of plasmonic nanoparticles	Tunable, passive, wavelength-specific light-to-heat conversion
Luminescent Downconversion	• Passive spectral conversion without energy input or heat generation • Tunable and scalable • Positive effect on plant biomass demonstrated in multiple experiments and on commercial scale	• Poor efficiency of spectral conversion; self-absorption losses • Poor long-term stability • Difficulty to emit blue/UV wavelengths • Potential toxicity of fluorophores	Passive, tunable spectral conversion to boost selected plant processes and consequently yield
Luminescent Upconversion	• Possibility to passively convert low-energy thermal radiation to higher energy PAR wavelengths • Possibility to access blue and UV portion of spectrum for emission	• Poor conversion efficiency, under diffuse solar radiation (typically seen for intense, focussed light sources) • Considerable research, optimization needed for technological applications	Passive thermal management and simultaneous boost in PAR wavelengths; Qualitative enhancement (colour/flavour/nutrient content) by conversion to blue/UV wavelengths

Figure 3. A summary of the proposed spectrum manipulation technologies for adaptation to greenhouses—the first column shows a simple schematic representing each technology; the second and third column list the main advantages and challenges for each technology in its current state, whereas the fourth column lists the characteristic possibilities presented by each technology if optimized and combined with greenhouse covering materials.

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4. General Challenges and Outlook

Passive spectrum manipulation, in particular spectrum conversion, for greenhouse applications has gained momentum in recent times, as evidenced by the growing number of recent scientific publications in these areas. To translate academic research into commercially viable technology, a rigorous, scientific understanding of the effects of such technology is required. Thus, both in experimental measurements and greenhouse trials, good reference situation measurements are needed to reliably separate light spectrum effects (i.e., spectral conversion) from light intensity effects (i.e., shading or diffusion). The greenhouse microclimate as well as plant growth and development processes are both affected by several parameters in complex ways: different crop species show different preferences for sunlight or shading; different species and even variants respond differently to the light spectrum composition; changes in the microclimate can further affect plant growth and development. Accounting for all the factors that a particular spectrum-manipulating intervention can affect by studying all the relevant control cases gives a holistic understanding of the possible consequences of that intervention, rather than a limited understanding of very specific effects on one particular crop (or variant) under very specific conditions. This broader scientific understanding of the effects of spectral manipulation on plant growth and development will benefit the field as a whole, providing a solid science-based foundation to assess the necessary case-by-case optimizations to attain the best possible outcomes in each specific case. Other beneficial experimental practices include: complete reporting of all physiological, photomorphic, quantitative, and qualitative results of a spectrum manipulation intervention; standardization in the reporting of measures like luminescence yield and quantification of spectral conversion; explicit clarification of all measurement conditions such as illumination and photon flux density with and without the intervention.

As the design of spectrum manipulating materials grows more complex—combining multilayers, periodic structures, or even more than one technology—scaling the fabrication up from laboratory samples to commercial greenhouses while maintaining nano/micrometer-scale precision will also present technical and economic challenges.

While the biggest advantages of passive technology lie in its simplicity, and lower maintenance and energy-use costs compared to active technology, a definite drawback is the inability to adapt to changing light and climate conditions, which are only expected to grow more sporadic with climate change. This could be solved to a limited extent by using such technology in screens, which can be deployed or withdrawn on demand. Another solution might lie in integrating some of these technologies with temporary or removable spray-on application.

Some guidelines for future studies and development can be laid down in terms of three main aspects: (i) crop productivity and quality, (ii) greenhouse microclimate, and (iii) sustainability.

In terms of crop productivity and quality, the most important spectrum manipulation is a boost in the relevant wavelengths, that is, blue and red, and, wherever required, UV: increases in leaf area, weight, and size of fruits have been well-established, but a more interesting goal would be increased secondary metabolite production to tailor color and flavor and to fortify nutrients.

In terms of the greenhouse microclimate, the most important manipulation concerns the NIR region of the spectrum—which strongly depends on the greenhouse location. In high-light-intensity tropical regions, the focus would be on NIR-blocking, such as through photonic crystals, or possibly upconversion. For temperate regions, technology that can retain heat is more beneficial, such as technology based on plasmonic materials. Another factor to choose the relevant substrates for incorporating spectral conversion elements is diffusion, which homogenizes the light distribution and microclimate within the greenhouse. Diffusion arises naturally in luminescent materials, which emit light isotropically, but in general other technology would benefit by being combined with diffusion, as long as this does not negatively impact PAR transmission.

Finally, in terms of sustainability, it is important to work on improving the long-term stability of such materials—both the durability of the substrate, and the stability of the active materials, for example, extending the lifetime of luminescent pigments. Sourcing or fabricating the active components of the spectrum manipulation technology as well as the substrates from earth-abundant, organic, bio-based, or easily recyclable materials is an important direction for research and development. Combining spectrum manipulating materials with spray-on technology could even do away with the need for substrates, minimizing waste generation over multiple cycles of application. Yet another important consideration is the development of materials that are free from toxic components such as heavy metals.

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Conflict of Interest

The authors declare no conflict of interest.

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fluorophores, greenhouses, light spectrum controls, luminescence, photonic crystals, plasmonics, quantum dots, sunlight

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