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<th>Title</th>
<th>Sharp selective scattering of red, green, and blue light achieved via gain material’s loss compensation</th>
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<tr>
<td>Author(s)</td>
<td>Ye, Yiyang; Liu, Rongyue; Song, Zhigang; Liu, Zhen; Chen, Tupei</td>
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</table>
Sharp selective scattering of red, green, and blue light achieved via gain material’s loss compensation

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Abstract: Frequency-selective scattering of light can be achieved by metallic nanoparticle’s localized surface plasmon resonance (LSPR). And this property may find an application in a transparent projection screen: ideally, specially designed metallic nanoparticles dispersed in a transparent matrix only selectively scatter red, green and blue light and transmit the visible light of other colors. However, optical absorption and surface dispersion of a metallic nanoparticle, whose size is comparable or smaller than mean free path of electrons in the constituent material, degenerate the desired performance by broadening the resonance peak width (i.e., decreasing frequency-selectivity) and decreasing light scattering intensity. In this work, it is shown that the problem can be solved by introducing gain material. Numerical simulations are performed on nanostructures based on silver (Ag), gold (Au) or aluminum (Al) with or without gain material, to examine the effect of gain material and to search for suitable structures for sharp selective scattering of red, green and blue light. And it is found that introducing gain material greatly improves performance of the structures based on Ag or Au except the structures based on Al. The most suitable structures for sharp selective scattering of red, green and blue light are, respectively, found to be the core-shell structures of silica/Au (core/shell), silica/Ag and Ag/silica, all with gain material.

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1. Introduction

Transparent screen is demanded for applications such as head-up display where navigation or other information is shown on windshield, shop window advertisement where vivid images or movies are displayed on window wall. Several solutions have been proposed to achieve the purpose of transparent screen, each having its own advantages and disadvantages. Micro-lens array based screen which uses beam splitter to redirect images into viewers’ eyes, has complicated structures to fabricate and limited viewing angle [1]. While a diffusive screen achieved by dispersing particles with high refractive index into a transparent matrix has a larger viewing angle, and is simple to fabricate [2], it suffers from trade-off between image clearness and screen’s transparency. Another well-researched screen is the organic light emitting diode (OLED) screen, consisting of OLED and transparent electrodes. Although OLED screen has high transparency and clear image view, it suffers from limited lifetime of its organic materials [3], and difficulty in scaling to large area. So, it is of interest to search for a way to overcome the shortages of existing solutions. Recently, transparent projection screens that selectively scatter red, green and blue light has been proposed based on metallic nanoparticle’s localized surface plasmon resonance (LSPR) [4–7], where in the ideal case metallic nanoparticles dispersed in a transparent matrix only selectively scatter red, green and blue light and transmit visible light of other colors (as shown in Fig. 1). This approach is
scalable to large sizes, can ensure image clearness and transparency by frequency-selective of light scattering, and has large viewing angle as will be demonstrated later in this manuscript.

Fig. 1. Schematic illustration of the concept of transparent projection screen achieved by selective scattering of red, green and blue light.

Nevertheless, due to metal’s optical loss, light absorption is inevitable in LSPR peak(s) of a metallic nanoparticle, which is undesirable for scattering-based applications. In addition, since a metallic nanoparticle’s size is comparable or smaller than mean free path of electrons in the constituent material, surface scattering of conduction electrons becomes significant [8–10], which undesirably increases light absorption and decreases the frequency-selectivity of light scattering by broadening the LSPR peak width [11–15]. Nevertheless, these problems may be solved by incorporating gain materials such as rare earth ions and laser dyes, which compensates optical loss and narrows LSPR peak width [16–25].

In this work, through numerical simulation, we aim to achieve sharp scattering peaks at red, green and blue light, with low absorption in the visible light range (400 nm ~800 nm) based on metallic nanoparticle’s LSPR effect and gain material’s loss compensation. Optimizations based on metallic nanosphere and core-shell structures of metal/silica (core/shell) and silica/metal are conducted for gold (Au), silver (Ag) and aluminium (Al), with and without gain material. During the optimizations, the effect of surface scattering of conduction electrons is considered. The results from the optimizations show that for the Ag- and Au-based structures, introducing gain material significantly improves the scattering behavior (i.e., there are sharper scattering peaks at red, green and blue light, respectively, and low absorption over visible light range), however the improvement is not significant for the Al-based structures. And it is found that the most suitable structures for sharp selective scattering at the red, green and blue light wavelength regions are silica/Au, Ag/silica and silica/Ag, respectively, all with a gain material.

2. Simulation method

In this section, reasons of choosing the metals and structures for subsequent optimizations are given, and the method of optimization is described.

Frequency-selectivity of light scattering is quantized by the width of a LSPR peak (the narrower, the better), and LSPR is a result of resonant oscillation of free electrons in response to incident electromagnetic wave [26,27]. Because of this, for metals to be employed in
subsequent simulations, their resonant peaks resulting from free electrons’ oscillations should be clearly separated from the broad spectrums caused by inter-band transitions of bound electrons [28] (i.e., if they overlap, then the peak width is broadened, resulting in a poor frequency-selectivity). According to this criteria, alkali metals, Al, Au and Ag are the candidates. However, alkali metals are excluded because they are very reactive. So, only Al, Au and Ag will be used in the following simulations.

The surrounding medium’s refractive index $n_s$ is assumed to be a constant of 1.5, which is about the same as the refractive indices of glass and most polymers, in which the nanoparticles to be designed are supposed to be dispersed. In quasi-static limit, namely, when the size of a metallic nanosphere is much smaller than the wavelength of incident light, the LSPR peak appears at a particular wavelength at which the real part of the metal’s dielectric function is equal to negative 2 times of surrounding medium’s dielectric function [29], i.e.,

$$\text{Re}\left[\varepsilon_m(\lambda_R)\right] = -2 \cdot \varepsilon_s$$

where $\lambda_R$ is the resonance peak wavelength and $\varepsilon_m(\lambda_R)$ is metal’s dielectric function at this wavelength, and $\varepsilon_s = n_s^2$ is surrounding medium’s dielectric function. For Ag, Au and Al, in quasi-static limit, their resonance peaks appear at 416 nm, 540 nm and 192 nm respectively, as observed from Fig. 2 below.

![Fig. 2. Calculated extinction efficiency spectrum of a single nanosphere with diameter of 10 nm by Mie theory [28], for (a) Ag, (b) Au and (c) Al. The real part of dielectric function is plotted for (d) Ag, (e) Au and (f) Al. The surrounding medium’s refractive index is 1.5, so surrounding medium’s dielectric function is $\varepsilon_s = n_s^2 = 2.25$, and the resonance peaks appear at the wavelengths at which $\text{Re}\left[\varepsilon_m(\lambda)\right] = -2 \cdot \varepsilon_s = -4.5$. The dielectric functions of the metals are from literature [30]. Definition of extinction efficiency is given in “Results and discussion”.

Increasing the metallic nanosphere’s size red-shifts its resonance peak, but further decreasing the size of a nanosphere with a size already much smaller than the wavelength of incident light does not significantly blue-shift its resonance peak [29]. Thus, here, the resonance peak wavelengths for Ag, Au and Al spheres in the quasi-static limit are perceived as the lower limits of their corresponding tuning ranges of resonance peak wavelength. The wavelengths of red, green and blue light are around 630 nm, 520 nm and 450 nm,
respectively. So, here, Au is considered only for red light scattering, as the lower limit (540 nm) of its resonance-peak-wavelength tuning range is larger than the wavelength of green light, while Ag and Al are considered for scattering of all three colors of light. The next step is to red-shift the metallic nanoparticles’ resonance peak wavelengths from their respective ones in the quasi-static limit, to red light for Au, and to all three colors of light for Ag and Al. A small red-shift of resonance peak of less than about 50 nm can be achieved by increasing the metallic nanospheres’ size. But relying on increasing size to red-shift resonance peak by more than 100 nm has the following problem: the dipole resonance peak’s strength will decrease, and the quadruple mode peak will appear and gradually become dominant, undesirably resulting in an overall broad extinction spectrum [29]. A feasible solution to achieve flexible red-shift of resonance peak is to use the structure of metallic nanoshell (i.e., dielectric/metal), which easily tunes the resonance peak wavelength via the ratio of shell thickness to core diameter [4,15,31]. Therefore, the structures of metallic nanosphere and nanoshell are employed in subsequent optimizations when gain material is not included, with the assumption that the dielectric core of nanoshell is silica. In the case of metallic nanoshell, the gain material is introduced by doping the silica core with gain material; while in the metallic nanosphere’s case, the gain material is introduced by surrounding the metallic nanosphere by a shell of silica doped with gain material. Experimental realization of doping silica shell of Au/Silica structure with gain material was demonstrated in a previous work [32].

With the above considerations, the structures to be employed during optimization are summarized in Table 1 below and are schematically depicted in Fig. 3.

![Schematic illustration of the structures to be used during optimization.](image)

Fig. 3. Schematic illustration of the structures to be used during optimization, where \( \varepsilon_m \) is the dielectric function of metal (Ag, Au or Al), \( \varepsilon_{Sil} \) is the dielectric function of silica (subject to whether or not doped with gain material, to be explained later), and \( \varepsilon_s = 2.25 \) is the dielectric function of the surrounding medium. (a) A single metallic nanosphere. (b) The core-shell structure of “Silica/Metal” without gain material. (c) The core-shell structure of “Metal/Silica”, silica shell is doped with gain material. (d) The core-shell structure of “Silica/Metal”, silica core is doped with gain material.
Table 1. Structures on which optimizations are to be performed to search for solutions of sharp selective scattering of red, green and blue light.

<table>
<thead>
<tr>
<th></th>
<th>Without gain material</th>
<th>With gain material</th>
</tr>
</thead>
<tbody>
<tr>
<td>Red</td>
<td>Au sphere, silica/Au, silica/Ag, silica/Al</td>
<td>Au/silica, silica/Au, silica/Ag, silica/Al</td>
</tr>
<tr>
<td>Green</td>
<td>Ag sphere, silica/Ag, silica/Al</td>
<td>Ag/silica, silica/Ag, silica/Al</td>
</tr>
<tr>
<td>Blue</td>
<td>Ag sphere, silica/Ag, silica/Al</td>
<td>Ag/silica, silica/Ag, silica/Al</td>
</tr>
</tbody>
</table>

For the purpose of optimization, a figure of merit ($FOM$) is defined:

$$FOM = \frac{\sigma_{sca}(\lambda_r)}{2\sigma_{sca} + \max\{\sigma_{abs}\}}$$

(1)

where $\sigma_{sca}$ and $\sigma_{abs}$ are the scattering and absorption cross sections, respectively, which are calculated by Mie theory [28], or the extension of the theory for core-shell structure [33]. The overbar and the symbol $\max\{\ldots\}$ denote the mean and the maximum values respectively in the visible spectrum (from 400 nm to 800 nm), and $\lambda_r$ is the resonance wavelength as mentioned earlier. This $FOM$ is defined in the way reported in reference [4], except the introduction of an absolute value operator on $\sigma_{abs}$. This $FOM$ favours the following properties: high scattering cross section at resonant wavelength and low scattering cross section elsewhere to form a sharp scattering peak and an overall small value of absorption cross section. Introducing gain material can make the absorption cross section negative, and eventually result in a lasing state [21] if the absolute value operator is not put on $\sigma_{abs}$ in Eq. (1) during optimization of $FOM$. The purpose of introducing gain material is to just compensate light absorption caused by metallic nanoparticle’s optical loss, and a low magnitude of overall absorption cross section is preferred over its mathematical value.

Since the size of a metallic nanoparticle is comparable to free electrons’ mean free path in the constituent material, effect of surface scattering of conduction electrons on the metal’s dielectric function cannot be ignored [8]. This issue is taken into account in Eq. (2) [34]:

$$\varepsilon_m(\omega) = \varepsilon_{exp}(\omega) + \frac{\omega_p^2}{\omega(\omega+i\gamma_s)} - \frac{\omega_p^2}{\omega(\omega+i(\gamma_b+i\gamma_s))}$$

(2)

where $\varepsilon_m$ is metal’s dielectric function corrected for size effect. In Eq. (2), $\omega$ is frequency of incident light, $\varepsilon_{exp}$ is the dielectric function of the bulk metal (Ag, Au or Al) obtained from literature [30], $\omega_p$ and $\gamma_b$ are the bulk metal’s plasma frequency and damping rate, respectively, whose values for Ag, Au and Al are given in Table 2 [34], $i$ is the imaginary number, $\gamma_s$ is the extra damping of free electrons caused by surface scattering. $\gamma_s$ in Eq. (2) is given by Eq. (3) [34]:

$$\gamma_s = \frac{V_F}{L_B}$$

(3)

where $V_F$ is the Fermi velocity of electrons in metal, and its values for Ag, Au and Al are given in Table 2 [34], and $L_B$ is the effective mean free path of free electrons in metallic
nanoparticle. For the structure of metallic nanoshell, i.e., silica/metal, $L_B$ is given by Eq. (4), derived by assuming specular reflections of electrons from surfaces of metallic shell [35]:

$$L_B = \frac{4(r_o^3 - r_i^3)}{3(r_o^3 + r_i^3)}$$  \tag{4}

where $r_o$ and $r_i$ are the outer and inner radii of the core-shell structure, respectively. For metallic nanosphere or the silica-coated metallic nanosphere (i.e., metal/silica), $L_B$ is given by Eq. (5), which is derived by setting $r_i$ to 0 in Eq. (4):

$$L_B = \frac{4r}{3}$$  \tag{5}

where $r$ is the radius of metallic nanosphere or the inner radius of silica-coated metallic nanosphere.

Table 2. Plasma frequencies, damping rates and Fermi velocities of bulk metals used in correcting the metals’ dielectric functions for the size effect [34].

<table>
<thead>
<tr>
<th></th>
<th>$\omega_p$ (eV)</th>
<th>$\gamma_p$ (eV)</th>
<th>$v_F$ ($10^6 m / s$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ag</td>
<td>9.6</td>
<td>0.0228</td>
<td>1.39</td>
</tr>
<tr>
<td>Au</td>
<td>8.55</td>
<td>0.0184</td>
<td>1.40</td>
</tr>
<tr>
<td>Al</td>
<td>15.3</td>
<td>0.5984</td>
<td>2.03</td>
</tr>
</tbody>
</table>

For silica without gain material, it is assumed to have a constant refractive index $N_{Sil} = 1.45$. For silica doped with gain material, its refractive index is given by [21]

$$N_{Sil} = 1.45 + ik$$  \tag{6}

where $i$ is the imaginary number, and $k$ is a negative extinction coefficient constant over all wavelengths, since reported gain materials are able to cover the whole visible wavelength range [16,18]. Extinction coefficient $k$ is positive for lossy media, 0 for non-absorbing media, and negative for gain media.

The optimization is carried out with BOBYQA [36], a local optimization algorithm, which starts from an initial guess of tuning parameters and searches for an optimized set of tuning parameters that give maximum value of $FOM$. Here, the set of tuning parameters are geometric parameters of the nanostructures shown in Table 1 (i.e., core diameter and shell thickness for metal/silica and silica/metal, or diameter for metal sphere), the value of $k$ in Eq. (6), and resonance peak wavelength $\lambda_R$. The tuning ranges of resonance peak wavelength $\lambda_R$ for red, green and blue light are set to be 620 nm to 700 nm, 510 nm to 540 nm, and 440 nm to 460 nm, respectively. The detailed optimization process is as following: for each tuning range of $\lambda_R$, $\lambda_R$ is scanned at a step of 1 nm; at each step of $\lambda_R$, BOBYQA is carried out with an initial guess of nanostructure’s dimensions and $k$ value; and the set that generates the largest value of $FOM$ defined by Eq. (1) gives the optimized $\lambda_R$, nanostructure’s dimensions and $k$ value. For structures without gain material, $k$ equals to 0 and is not an adjustable parameter during optimization. The optimized results of nanostructure’s
dimensions, $k$ values and values of optimized $FOM$ are shown in the section of “Results and discussion.”

3. Results and discussion

In this section, apart from the optimized tuning parameters, and values of optimized $FOM$, the calculated optical spectrums are also presented to give an intuitive view of sharp selective scattering.

The spectrums are presented in terms of extinction efficiency ($Q_{\text{ext}}$), scattering efficiency ($Q_{\text{sca}}$) and absorption efficiency ($Q_{\text{abs}}$), which are obtained from dividing their corresponding cross sections: $\sigma_{\text{ext}}$, $\sigma_{\text{sca}}$ and $\sigma_{\text{abs}}$, calculated by Mie theory [28,33], to the geometrical cross section $\pi R^2$, where $R$ is the overall radius of a core-shell structure (metal/silica or silica/metal) or radius of a nanosphere, i.e., $Q_{\text{ext}} = \sigma_{\text{ext}} / (\pi R^2)$, $Q_{\text{sca}} = \sigma_{\text{sca}} / (\pi R^2)$, and $Q_{\text{abs}} = \sigma_{\text{abs}} / (\pi R^2)$. Note that $Q_{\text{ext}}$ is equal to the sum of $\sigma_{\text{sca}}$ and $\sigma_{\text{abs}}$.

For the calculated efficiencies shown in Figs. 4, 5 and 6, Mie theory takes the following parameters as its inputs: the surrounding medium’s dielectric function $\varepsilon_s = n_s^2 = 2.25$, optimized nanostructure’s dimensions (i.e., core diameter and shell thickness for a core-shell structure, or diameter for a nanosphere), metal’s size-dependent dielectric function given by Eq. (2), silica’s dielectric function $\varepsilon_{\text{Sil}}$ for the core-shell structure (for a structure without gain material, $\varepsilon_{\text{Sil}} = N_{\text{Sil}}^2 = 2.1025$; for a structure with gain material $\varepsilon_{\text{Sil}} = N_{\text{Sil}}^2 = (1.45 + ik)^2$, where $N_{\text{Sil}} = 1.45 + ik$ is from Eq. (6) and the optimized $k$ is constant over the whole spectrum).

Figure 4 shows the efficiency spectrums of the structures optimized for selective scattering of red light, and their corresponding values of $FOM$ as well as optimized parameters are shown in Table 3.

<table>
<thead>
<tr>
<th>Alphabetical order</th>
<th>Core Diameter (nm)</th>
<th>Shell thickness (nm)</th>
<th>$k$</th>
<th>Resonance wavelength (nm)</th>
<th>$FOM$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>101</td>
<td></td>
<td></td>
<td>620</td>
<td>0.7476</td>
</tr>
<tr>
<td>(b)</td>
<td>98</td>
<td>5</td>
<td>0.4663</td>
<td>620</td>
<td>1.1094</td>
</tr>
<tr>
<td>(c)</td>
<td>61</td>
<td>23</td>
<td></td>
<td>650</td>
<td>0.7640</td>
</tr>
<tr>
<td>(d)</td>
<td>48</td>
<td>10</td>
<td>1.2888</td>
<td>622</td>
<td>3.17</td>
</tr>
<tr>
<td>(e)</td>
<td>65</td>
<td>18</td>
<td></td>
<td>620</td>
<td>0.6063</td>
</tr>
<tr>
<td>(f)</td>
<td>56</td>
<td>8</td>
<td>1.1234</td>
<td>620</td>
<td>1.3391</td>
</tr>
<tr>
<td>(g)</td>
<td>97</td>
<td>25</td>
<td></td>
<td>672</td>
<td>0.4509</td>
</tr>
<tr>
<td>(h)</td>
<td>83</td>
<td>25</td>
<td>0.0677</td>
<td>622</td>
<td>0.4853</td>
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</tbody>
</table>

Figure 5 shows the efficiency spectrums of the structures optimized for selective scattering of green light, and their corresponding value of $FOM$ as well as optimized parameters are shown in Table 4.
Table 4. Optimized tuning parameters and values of optimized $FOM$ for the structures mentioned in Fig. 5, with the same alphabetic order

<table>
<thead>
<tr>
<th>Alphabetic order</th>
<th>Core Diameter (nm)</th>
<th>Shell thickness (nm)</th>
<th>$k$</th>
<th>Resonance wavelength (nm)</th>
<th>$FOM$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>78</td>
<td>510</td>
<td></td>
<td></td>
<td>0.8163</td>
</tr>
<tr>
<td>(b)</td>
<td>78</td>
<td>15</td>
<td>−0.1743</td>
<td>510</td>
<td>1.1289</td>
</tr>
<tr>
<td>(c)</td>
<td>20</td>
<td>29</td>
<td></td>
<td></td>
<td>0.8087</td>
</tr>
<tr>
<td>(d)</td>
<td>34</td>
<td>9</td>
<td>−1.2435</td>
<td>511</td>
<td>2.4128</td>
</tr>
<tr>
<td>(e)</td>
<td>65</td>
<td>18</td>
<td></td>
<td></td>
<td>0.6329</td>
</tr>
<tr>
<td>(f)</td>
<td>62</td>
<td>18</td>
<td>−0.8485</td>
<td>510</td>
<td>0.6522</td>
</tr>
</tbody>
</table>

Figure 6 shows the efficiency spectrums of the structures optimized for selective scattering of green light, and their corresponding value of $FOM$ as well as optimized parameters are shown in Table 5.

Table 5. Optimized tuning parameters and values of optimized $FOM$ for the structures mentioned in Fig. 6, with the same alphabetic order

<table>
<thead>
<tr>
<th>Alphabetic order</th>
<th>Core Diameter (nm)</th>
<th>Shell thickness (nm)</th>
<th>$k$</th>
<th>Resonance wavelength (nm)</th>
<th>$FOM$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(a)</td>
<td>67</td>
<td>460</td>
<td></td>
<td></td>
<td>0.7123</td>
</tr>
<tr>
<td>(b)</td>
<td>47</td>
<td>19</td>
<td>−0.1726</td>
<td>443</td>
<td>2.5227</td>
</tr>
<tr>
<td>(c)</td>
<td>17</td>
<td>24</td>
<td></td>
<td></td>
<td>0.6569</td>
</tr>
<tr>
<td>(d)</td>
<td>18</td>
<td>27</td>
<td>−1.44</td>
<td>460</td>
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</tr>
<tr>
<td>(e)</td>
<td>31</td>
<td>23</td>
<td></td>
<td></td>
<td>0.9602</td>
</tr>
<tr>
<td>(f)</td>
<td>27</td>
<td>26</td>
<td>−0.8688</td>
<td>440</td>
<td>0.9794</td>
</tr>
</tbody>
</table>
Fig. 4. Efficiency spectrums of optimized structures for selective scattering of red light. The left column, i.e., (a), (c), (e) and (g) are for the optimized structures without gain material, and spectrums of their corresponding structures optimized with gain material are shown in the right column, i.e., (b), (d), (f) and (h). (a) Au sphere without gain. (b) Au/silica with gain. (c) Silica/Au without gain. (d) Silica/Au with gain. (e) Silica/Ag without gain. (f) Silica/Ag with gain. (g) Silica/Al without gain. (h) Silica/Al with gain. For each structure, the optimized parameters and optimized values of $FOM$ are tabulated in Table 3 in the same alphabetic order as their respective structure appears in this figure.
Fig. 5. Efficiency spectrums of optimized structures for selective scattering of green light. The left column, i.e., (a), (c) and (e) are for the optimized structures without gain material, and spectrums of their corresponding structures optimized with gain material are shown in the right column, i.e., (b), (d) and (f). (a) Ag sphere without gain. (b) Ag/silica with gain. (c) Silica/Ag without gain. (d) Silica/Ag with gain. (e) Silica/Al without gain. (f) Silica/Al with gain. For each structure, the optimized parameters and optimized values of $FOM$ are tabulated in Table 4 in the same alphabetic order as their respective structure appears in this figure.
Fig. 6. Efficiency spectrums of optimized structures for selective scattering of blue light. The left column, i.e., (a), (c) and (e) are the optimized structures without gain material, and spectrums of their corresponding structures optimized with gain material are shown in the right panels, i.e., (b), (d) and (f). (a) Ag sphere without gain. (b) Ag/silica with gain. (c) Silica/Ag without gain. (d) Silica/Ag with gain. (e) Silica/Al without gain. (f) Silica/Al with gain. For each structure, the optimized parameters and optimized values of $FOM$ are tabulated in Table 5 in the same alphabetic order as their respective structure appears in this figure.
Negative absorption is observed for some structures, which is interpreted as radiation of light. The occurrence of negative absorption is reasonable with the introduction of gain material, as gain material is used in lasers to achieve lasing state which is a state of light radiation.

By observing Figs. 4, 5 and 6, it is obvious that Al-based structures are not suitable for selective scattering, since their resonance peaks are too broad, even though absorptions are low. The efficiency spectrums for Al-based structures do not show apparent improvements even after gain material is introduced. And the broad resonance peaks for Al-based structures can be explained by Al’s low quality factor, which determines strength of resonance, defined by $-\text{Re} \left[ \varepsilon_n \right] / \text{Im} \left[ \varepsilon_n \right]$, over the whole visible wavelength range [34].

Apart from structures of Al, by comparing the calculated efficiency spectrums of the structures without gain material to those of structures with gain material in Figs. 4, 5 and 6, it is observed that introducing gain material significantly improves the scattering quality in the sense that the ratios of absorption are decreased, and the resonance scattering peaks become sharper. And the improvements after introducing gain material are confirmed by the increase of values of $FOM$ as shown in Tables 3, 4 and 5.

The largest values of $FOM$ in Tables 3, 4 and 5 (3.17, 2.4128 and 2.5227 respectively) suggest that the most suitable structures for sharp selective scattering of red, green and blue light are silica/Au with gain material (see Table 3), silica/Ag with gain material (see Table 4), and Ag/silica with gain material (see Table 5), respectively. Their angular distributions of scattering are shown in Fig. 7. The results shown in Fig. 7 suggest that the scattered light can be viewed from a wide range of angle. The $FOM$ values of the most suitable structures are much larger than those reported in previous works [4,6,7], as can be seen from Table 6, showing a great improvement in selective scattering of red, green and blue light.
Fig. 7. Intensity of the scattered light at the resonant wavelengths versus the scattering angle (in the unit of degree) in polar form for the most suitable structures respectively for selective scattering of red, green and blue light: (a) Silica/Au with gain material (Table 3), (b) Silica/Ag with gain material (Table 4), and (c) Ag/silica with gain material (Table 5). The incident light has a unit intensity and equal intensity for the p- and s- components with respect to the scattering plane. Calculations are carried out with the Mie theory.

Table 6. Comparison of optimized values of FOM from this work to those from previous works for red, green and blue light

<table>
<thead>
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<th></th>
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<th>Green</th>
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Among the three most suitable structures, the one for selective scattering of red light, i.e., the structure of silica/Au with gain material (item d in Table 3), has the largest magnitude of optimized $k$ which is 1.2888, and this corresponds to a gain value of $2.6 \times 10^5 \text{cm}^{-1}$, according to the relation $gain = -\frac{4\pi k}{\lambda}$ [16], where $\lambda = 622\text{nm}$ is this structure’s optimized resonance wavelength. The relation between gain and the emission cross section $\sigma_e$ of a single gain material (dye or rare earth ion) is: $gain = \rho \cdot \sigma_e$ [19], where $\rho$ is gain material’s volume concentration. The usually-known gain material’s emission cross section is of the order of $10^{-16} \text{cm}^2$ [16,17,19], which corresponds to a maximum gain of the order of...
$10^3 \text{ cm}^{-1}$. However, it has been pointed out that the theoretical limit of emission cross section set by quantum mechanics is $\frac{3 \lambda^2}{(2 \pi)}$ \cite{37}, which is of the order of $10^{-10} \text{ cm}^2$ for visible wavelength range, and is 6 orders larger than $10^{-16} \text{ cm}^2$. So, a gain value of the order of $10^5 \text{ cm}^{-1}$ may be achieved theoretically.

![Fig. 8. Simulated transmittance of a film dispersed with the three most suitable structures.](image)

The transmittance spectrum of a film dispersed with the three most suitable structures is shown in Fig. 8, and details of simulating this transmittance is given in the following. According to Beer-Lambert law, transmittance $T(\lambda)$ of incident light of wavelength $\lambda$ after it travels a distance $t$ in a medium dispersed with nanoparticles is given by \cite{29}:

$$T(\lambda) = e^{-N \sigma_{\text{ext}}(\lambda) t} \quad (7)$$

In Eq. (7) $\sigma_{\text{ext}}(\lambda)$ is the extinction cross section of the dispersed nanoparticle (assuming only one kind of nanoparticle is dispersed) at a wavelength of $\lambda$, $N$ is the nanoparticle’s volume density. The product of $N$ and $t$ gives areal density $N’$ of the dispersed nanoparticle (the surface of this area is perpendicular to the propagation direction of incident light), and Eq. (7) can be written as:

$$T(\lambda) = e^{-N’ \sigma_{\text{ext}}(\lambda)} \quad (8)$$

The area densities of the three most suitable structures are chosen such that transmittance at their corresponding resonance wavelengths become 20%. And this results in an areal density of $1.90 \times 10^9 / \text{ cm}^2$ for red light scatterer (silica/Au with gain, or item d in Table 3), $3.06 \times 10^9 / \text{ cm}^2$ for green light scatterer (silica/Ag with gain, or item d in Table 4) and $4.47 \times 10^9 / \text{ cm}^2$ for blue light scatterer (Ag/silica with gain, or item b in Table 5). And the overall transmittance is the product of the three transmittance spectrums for the three scatterers.
4. Conclusion

In this work, for the purpose of realizing a transparent projection screen, we have searched for structures that may achieve sharp selective scattering of red, green and blue light based on metallic nanoparticle’s LSPR effect and gain material’s optical compensation. By comparing behaviors of structures optimized with gain material to those without gain material, it is found that for the structures based on Au and Ag, introducing gain material greatly improves the performance in terms of sharper scattering peak and overall lower absorption, while for the Al-based structures little improvement is observed. The most suitable structures for sharp selective scattering of red, green and blue light are respectively found to be silica/Au, silica/Ag and Ag/silica, all with gain material, with their corresponding optimized parameters shown in Tables 3, 4 and 5. And among gain values of the three most suitable structures, the largest one is still within physical limit.

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References

27. C. F. Bohren and D. R. Huffman, Absorption and scattering of light by small particles (John Wiley & Sons, 2008).