Controlled light scattering of a single nanoparticle by wavefront shaping

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Controlling light scattering by nanoparticles is fundamentally important for the understanding and the control of light inside photonic nanostructures, as well as for nanoparticle scattering itself, including Mie scattering. Here, we theoretically and numerically investigate the possibility to manipulate nanoparticle scattering through wavefront shaping that was initially developed to control scattering of light through opaque random media that consist of large numbers of scattering nanoparticles. We find that even a single nanoparticle supports multiple strongly scattering eigenchannels, which opens the opportunity to manipulate scattering with wavefront shaping previously developed for multiple scattered light through opaque random media. We find that these scattering eigenchannels are related to different resonant leaky modes of the scatterer. Moreover, we investigate the spectral correlation of these highly scattering eigenchannels, and demonstrate the coexistence of short range and long range correlations. Our work proposes a new tool to control light-matter interactions with resonant modes via wavefront shaping and constitutes a step towards exploring novel spectral correlations in the scattering of light by nano scatterers, including Mie spheres.

I. INTRODUCTION

The manipulation of waves, such as electronic waves, acoustic waves, and optical waves, is an ongoing central topic in many research fields [1]. In optics, many different mesoscopic nanostructures have been developed to control light waves, including photonic crystals and scattering media [2-4], plasmonic structures [5], metamaterials [6], and metasurfaces [7]. In particular, photonic scattering media that consist of many randomly-distributed nanoparticles, have been applied to control light propagation and light-matter interactions for, notably, Anderson localization and transverse localization [8-12], weak localization and coherent backscattering [13, 14], random lasing [15, 16], imaging through opaque media [17, 18], and sensitive deep inside opaque media such as biological tissue [19, 20]. Recently, optical wavefront shaping (WFS) [21, 22] has been demonstrated to be a powerful method to manipulate light in and through complex scattering media, by spatially shaping the incident wavefront, leading to novel applications in high-resolution imaging [23, 24], enhanced energy delivery [25, 26], efficient light emission [27, 28], and classical and quantum communication schemes [29, 30].

Many classes of nanostructures such as photonic crystals, metasurfaces, and scattering media, consist of assemblies of nanoscalers as the elementary structural motifs, hence light scattering plays a fundamental role in these nanostructures [31, 32]. Consequently, manipulation of light scattering is crucial for the understanding and control of light propagation in these complex nanostructures. To control the scattering of a single nanoparticle, typically the structural parameters are engineered, notably to tailor multipolar interference and scattering properties of the scatterer upon illumination by incident plane waves [33, 34]. When achieving resonant excitation for a nanoparticle, the excited cavity mode has been exploited to enhance light-matter and nonlinear interactions, such as, photoluminescence [35, 36], lasing [37, 38], and optical harmonic generation [39, 40]. Interestingly, it has been found that Mie resonances can be excited more efficiently with structured incident light [41, 42]. In this context, a little explored question is whether the scattering of even a single nanoparticle can also be manipulated through optical WFS?

![FIG. 1. Schematic diagram of the physical situation studied here. Incident light with a shaped wavefront is incident on a scattering particle (MS), which results in scattered waves in far field with controllable properties. This WFS diagram for manipulating Mie scattering can be realized practically, where the light scattered at an angle $\theta$ is monitored by a detector in the far field.](image-url)
optical interference between multiple light paths can be controlled by WFS [24, 25, 56]. Remarkably, from our analysis in this paper of the backscattering matrix of a single scatterer that is experimentally accessible, we find that there are also multiple strongly scattering eigenchannels for such a single scatterer. Therefore, optical WFS can effectively be employed to manipulate light scattering by selectively exciting these single-particle eigenchannels. The physical essence is the realization that light waves interacting with a single nanoparticle are not necessarily experiencing single scattering (in which case wavefront shaping control would not be feasible), but the waves are multiply scattered by the single nanoparticle, as described by the $t$-matrix [57] of a single scatterer [50, 58, 59].

By investigating the scattering light field inside the nanoparticle, we find that these highly scattering eigenchannels are related to different leaky resonances of the scatterer. Consequently, by employing optical WFS to selectively excite different scattering eigenchannels, it is possible to control the leakage enhanced light-matter interaction with individual nanoparticles. Moreover, we have demonstrated that the highly scattering eigenchannels exhibit both short range and long range spectral correlations, which is related to the rich resonant features of the nanoparticle. These results offer new perspectives for understanding and controlling the intriguing light scattering process in various nanostructures.

II. METHODS AND PRINCIPLE

The nanoparticle scattering diagram is shown in Fig. 1, where the wavefronts of the incident light are controlled by optical WFS. The nanoparticle scatters light into different angles in the far field, while we focus on the backscattering light field experimentally measurable with a detector. To thoroughly analyze the backscattering, we employ the random matrix theory to describe the relationship of the incident and outgoing free modes at different far-field angles [25, 60, 62], demonstrating the existence of multiple scattering eigenchannels with merely a single scatter. Notably, our work is also relevant to the outgoing light in the forward direction, where the field is a sum of the forward scattered fields and the incident field that may give rise to intriguing interferences. The existence of multiple highly scattering eigenchannels may shed new light on the recently reported scattering phenomena of mutual extinction and transparency [63, 64] for various nanoparticles and nanostructures.

Following the random matrix theory [25, 60, 62], we model the incident and outgoing free modes as column vectors, and obtain a backscattering matrix $S_r$ for the single nanoparticle. Specifically, the incident and outgoing free modes are represented by the column vectors $L_{\text{in}}$ and $L_{\text{out}}$, respectively. Meanwhile, we assume that the square of the $i$th element of $L_{\text{in}}^\ast L_{\text{in}}$ (or $L_{\text{out}}^\ast L_{\text{out}}$) denotes the power of light in the relevant incident (or scattering) channel. Hence, the $i$th element is related to the electric field by $|L_{\text{in}}^\ast|^2 = |E_{\text{in}}^\ast|^2 \cdot \Delta S_{\text{in}}$ (or $|L_{\text{out}}^\ast|^2 = |E_{\text{out}}^\ast|^2 \cdot \Delta S_{\text{out}}$), where $\Delta S_{\text{in}}$ (or $\Delta S_{\text{out}}$) is a constant value dependent on the area of the discrete incident (or scattering) channels. Once the scattering matrix $S_r$ is retrieved, for instance from interferometry, the backscattering by a single nanoparticle is obtained from the following expression

$$L_{\text{out}} = S_r L_{\text{in}}.$$  

(1)

Interestingly, one can employ singular value decomposition on $S_r$ to reveal the scattering eigenchannels, given by

$$S_r = USV^*,$$  

(2)

where $U$ and $V$ are unitary matrices, and $S$ is a diagonal matrix. Here, the eigenvalues of the backscattering channels are the diagonal elements of $S_r$ labeled as $s_i$, while the columns of the unitary matrix $V (\mathbf{U})$ represent the eigenfunctions (that is, wavefunctions) for the incident (scattering) spatial channels. Therefore, the distribution of the diagonal elements $s_i$ holds information of the scattering features of the single scatterer. Next, we will show, both by theory and full wave calculations, that with wavefront shaping we are indeed able to manipulate the scattering of a single nanoparticle. In other words, from the singular value decomposition, we find that a single scatterer supports many eigenchannels, of which a subset will appear to be strongly scattering.

III. SCATTERING EIGENCHANNELS OF A SINGLE SCATTERER

A. Nanoparticle in 2 dimensions

Configuration. We first consider a 2D nanoparticle with a rectangular shape, since this is straightforward to discretize on a rectangular computational grid. The height of the scatterer (along y axis) is $a = 300$ nm, and the width (along x axis) is $b = 600$ nm. The refractive index of the scatterer material is taken to be $n = 3.5$, typical of high-index semiconductors like Si or GaAs [65, 66]. The backscattering matrix $S_r$ is obtained by sequentially sending plane incident waves at different incident angles and simultaneously monitoring the scattered field in the far field plane. In our full wave simulation, a z-polarized incident light $E_{\text{inc}}(\theta_{\text{in}})$ is scanned from $-45^\circ$ to $45^\circ$ (anticlockwise from $\hat{y}$) with a stepsize $\Delta\theta_{\text{in}} (=1^\circ)$. This scanning range corresponds to a numerical aperture $NA = 1/\sqrt{2} \approx 0.71$, which is practically realizable with a commercial objective lens. Meanwhile, the backscattered field $E_{\text{back}}(\theta_{\text{out}})$ with $\theta_{\text{out}} \in [-45^\circ, 45^\circ]$ (anticlockwise from $-\hat{y}$) is obtained at a sampling step $\Delta\theta_{\text{out}} (=1^\circ)$, which is easily measurable in practice with the same objective lens used for sending the incident light.

Methods. With the above configuration, the incident and backscattered electric field is described by a column

...
vector \( \mathbf{E}_{\text{in}} \) (\( \mathbf{E}_{\text{out}} \)) with \( N \) (\( = 91 \)) elements, each element of which is the electric field at a specific angle. The backscattered light field is related to the incident light field by employing a scattering matrix \( \mathbf{M} \) with dimensions \( N \times N \) (here \( N = 91 \)), expressed as

\[
\mathbf{E}_{\text{out}} = \mathbf{M} \mathbf{E}_{\text{in}}.
\]  

By sequentially scanning the angle of incident light and monitoring the backscattered light, we obtain the scattering matrix \( \mathbf{M} \). To further relate \( \mathbf{M} \) to power-related backscattering matrix \( \mathbf{S}_{\text{scat}} \), we need to calculate the power of each incident channel against the light field, as well as that of the backscattering channels. For \( i \)th incident channels, the power of light illuminating on the scatterer is expressed as

\[
P_{\text{in}}^i = |\mathbf{E}_{\text{in}}^i|^2 = |E_{\text{in}}^i|^2 \cdot \sigma_{\text{scat}}/\Delta \theta_{\text{in}},
\]

where \( E_{\text{in}}^i \) is the electric field of the incident plane wave from the \( i \)th-channel, and \( \sigma_{\text{scat}} \) denotes the cross size of the scatterer multiplied by a unit length of \( z \) axis. For a subwavelength scatterer, the value of \( \sigma_{\text{scat}} \) could be set as the length of Airy spot \( \lambda/n_1 \) in 2D. Here, \( \lambda \) is the wavelength, and \( n_1 \) is the refractive index of the material surrounding the scatterer. Then, we derive to obtain the column vector \( \mathbf{L}_{\text{in}} \) as

\[
\mathbf{L}_{\text{in}} = \mathbf{E}_{\text{in}} \cdot \sqrt{\sigma_{\text{scat}}/\Delta \theta_{\text{in}}}.
\]  

The vector \( \mathbf{L}_{\text{in}} \) is related to the total input power \( P_{\text{in}}^\text{tot} \) as

\[
P_{\text{in}}^\text{tot} = \mathbf{L}_{\text{in}}^\text{tot} \mathbf{L}_{\text{in}}.
\]

By considering the cylindrical surface of the far-field scattering light, the power \( P_{\text{out}} \) of scattered light is expressed as

\[
P_{\text{out}}^j = |\mathbf{E}_{\text{out}}^j|^2 = |E_{\text{out}}^j|^2 \cdot R\Delta \theta_{\text{out}},
\]

where \( R \) is radius of the far-field circle. Then, we derive to obtain the column vector \( \mathbf{L}_{\text{out}} \) as

\[
\mathbf{L}_{\text{out}} = \mathbf{E}_{\text{out}} \cdot \sqrt{R \Delta \theta_{\text{out}}}.
\]  

The matrix \( \mathbf{L}_{\text{out}} \) is related to the total output power \( P_{\text{out}}^\text{tot} \) as

\[
P_{\text{out}}^\text{tot} = \mathbf{L}_{\text{out}}^\text{tot} \mathbf{L}_{\text{out}}.
\]

Now, by substituting Eqs. 5 and 6 to Eq. 1, the backscattering matrix \( \mathbf{S}_{\text{scat}} \) is derived to be

\[
\mathbf{S}_{\text{scat}} = \beta_{2D} \cdot \mathcal{M}
\]

where the constant prefactor is equal to \( \beta_{2D} = \sqrt{R \Delta \theta_{\text{out}} \Delta \theta_{\text{in}}}/\sigma_{\text{scat}} \).

Results. With the backscattering matrix \( \mathbf{S}_{\text{scat}} \), we carried out the singular value decomposition by using Eq. 2. We find that the nanoparticle supports \( N \) eigenchannels, of which a subset \( N_s \) appears to be strongly scattering. To quantify strongly scattering, we propose as a criterion to relate eigenvalues of higher order channels to the first, strongest, one \((s_j/s_1)\) and take as a gauge that the \( j \)th channel exceeds \((s_j/s_1 > 10\%)\) compared to the strongest one.

<table>
<thead>
<tr>
<th>( b/\lambda )</th>
<th>1/4</th>
<th>2/7</th>
<th>1/3</th>
<th>2/5</th>
<th>1/2</th>
<th>3/5</th>
<th>3/4</th>
<th>1</th>
</tr>
</thead>
<tbody>
<tr>
<td>( s_1 )</td>
<td>0.520</td>
<td>0.527</td>
<td>0.586</td>
<td>0.686</td>
<td>0.667</td>
<td>0.267</td>
<td>0.603</td>
<td>0.830</td>
</tr>
<tr>
<td>( s_2 )</td>
<td>0.158</td>
<td>0.129</td>
<td>0.135</td>
<td>0.350</td>
<td>0.356</td>
<td>0.216</td>
<td>0.411</td>
<td>0.414</td>
</tr>
<tr>
<td>( s_3 )</td>
<td>0.001</td>
<td>0.003</td>
<td>0.015</td>
<td>0.012</td>
<td>0.033</td>
<td>0.060</td>
<td>0.371</td>
<td>0.298</td>
</tr>
</tbody>
</table>

TABLE I. The eigenvalues of the first three highest scattering eigenchannels of the backscattering matrix at different frequencies. The results show that the subwavelength scatterer has \( N_s > 1 \) strongly scattering eigenchannels.

In Table I, we list the first three eigenchannels with substantial eigenvalues of the backscattering matrix with increasing frequency. Here, we consider the wavelength range with the incident wavelength larger than the scatterer’s dimension \( b \). At the lowest frequencies \( b/\lambda = 1/4, 2/7, 1/3, 2/5, 1/2 \), the \((j = 2)\) eigenvalue is strong, and the \((j = 3)\) eigenvalue is weakly scattering. Thus, the number of strongly scattering channels is \( N_s = 2 \). At the higher frequencies \( b/\lambda = 3/5, 3/4, 1 \), both the \((j = 2)\) and \((j = 3)\) eigenvalues are strongly scattering, thus \( N_s = 3 \). It is thus clear that the number of eigenchannels is greater than one \((N_s > 1, \text{as naively expected in the introduction})\), in other words, more than one highly scattering eigenchannels is sustained by the nanoparticle. By further increasing the size of the scatterer (or equivalently, increasing the frequency of incident light), more highly scattering channels are expected to appear. These results demonstrate that manipulation of nanoparticle scattering is feasible with optical WFS, by selectively addressing these different strongly scattering eigenchannels.

B. Mie sphere in 3 dimensions

Configuration. Generally, a full-wave simulation for an arbitrary 3D nanoparticle, including a Mie sphere, will require computationally a more intensive scan of the incident beam than in 2D, which is thus much more time consuming. Therefore, we focused on the Mie sphere that has analytical solutions on the scattering light [67, 68]. Without losing generality, we did theoretical calculations for a Mie sphere with diameter \( d_s = 400 \text{ nm} \) and refractive index \( n = 2 \), typical for a high-index material such as silicon nitride. The normalized frequency of incident light is taken to be \( d_s/\lambda = 8/9 \). Again, we consider the backscattering configuration, in which the incident angle of light \( \theta_{\text{in}} \in [0^\circ, 32^\circ] \) (relative to \( \hat{y} \)), and \( \phi_{\text{in}} \in [0^\circ, 360^\circ] \) (relative to \( \hat{x} \) in the \( xz \) plane). For the backscattered light, we have \( \theta_{\text{out}} \in [148^\circ, 180^\circ] \), and \( \phi_{\text{out}} \in [0^\circ, 360^\circ] \). This setting can be realized with a microscopic setup with numerical aperture \( NA = \sin(32^\circ) = 0.53 \). Subsequently, the electric field of the backscattered light \( \mathbf{E}_{\text{far}}(\theta_{\text{out}}, \phi_{\text{out}}) \) is calculated analytically in a far-field spherical surface of large radius \( R (= 1 \text{ m}) \), with an incident light \( \mathbf{E}_{\text{inc}}(\theta_{\text{in}}, \phi_{\text{in}}) \). To obtain the backscattering matrix \( \mathbf{S}_{\text{scat}} \), we sample \( \theta_{\text{in}} \) with \( N \) steps, and \( \phi_{\text{in}} \) with \( M \).
steps, and consequently the size of $E_{\text{in}}$ is $N_{\text{in}} = 2NM \times 1$, where the factor 2 is introduced due to the two independent polarization of the light field. For simplicity, we sample the backscattered light in a similar way as the incident field, and the size of $E_{\text{out}}$ is also $N_{\text{out}} = 2NM \times 1$. Here, the sampling steps $N$ is 8, and $M$ is 72. Therefore, the size of $L_{\text{in}}$ and $L_{\text{out}}$ is 1152, and the size of $S_r$ is $1152 \times 1152$.

Methods. By continuously scanning the angle of incident light and monitoring the backscattered light, we get a scattering matrix $M$ that relates the scattering field vector $E_{\text{out}}$ to the incident field vector $E_{\text{in}}$, i.e.,

$$E_{\text{out}} = \mathcal{M} E_{\text{in}},$$

(9)

Similarly as in the 2D case, we calculate the power-related backscattering matrix $S_r$. The power of $i$th input channel that illuminates on the scatterer is given by

$$P_{\text{in}}^i \equiv |E_{\text{in}}^i|^2 = |E_{\text{in}}^i|^2 \cdot \sigma_{\text{scat}} / (\Delta \theta_{\text{in}} \Delta \phi_{\text{in}}),$$

(10)

where $\sigma_{\text{scat}}$ denotes the cross section of the Mie sphere, and could be set as the area of air spot $\pi(0.61l_1)^2$ for a subwavelength scatterer. Here $\lambda$ is the wavelength, and $n_1$ is the refractive index of the environment material surrounding the scatterer. Then, we obtain the column vector $L_{\text{in}}$ as

$$L_{\text{in}} \equiv E_{\text{in}} \sqrt{\sigma_{\text{scat}} / (\Delta \theta_{\text{in}} \Delta \phi_{\text{in}})}.$$

(11)

The vector $L_{\text{in}}$ is related to the total input power $P_{\text{in}}^{\text{tot}}$ as $P_{\text{in}}^{\text{tot}} = L_{\text{in}}^\top L_{\text{in}}$. For the scattered light, the power of $j$th scattering channel at the far-field sphere surface is expressed as

$$P_{\text{out}}^j = |E_{\text{out}}^j|^2 \cdot R^2 \sin(\theta_{\text{out}}^j) \Delta \theta_{\text{out}} \Delta \phi_{\text{out}},$$

(12)

We obtain the column vector $L_{\text{out}}$ as

$$L_{\text{out}} = G(\theta_{\text{out}}^j) E_{\text{out}} \cdot R \sqrt{\Delta \theta_{\text{out}} \Delta \phi_{\text{out}}},$$

(13)

Here, $G(\theta_{\text{out}}^j)$ is a $N_{\text{out}} \times N_{\text{out}}$ matrix with the value of elements at $\theta_{\text{out}}^j$ being $\sqrt{\sin(\theta_{\text{out}}^j)}$. The matrix $L_{\text{out}}$ is related to the total output power $P_{\text{out}}^{\text{tot}}$ as $P_{\text{out}}^{\text{tot}} = L_{\text{out}}^\top L_{\text{out}}$. Now, by substituting Eqs. (11) and (13) to Eq. (11) the backscattering matrix $S_r$ is derived to be

$$S_r = \beta_{3D} \cdot G(\theta_{\text{out}}) \mathcal{M}$$

(14)

where the constant prefactor is equal to $\beta_{3D} = R \sqrt{\Delta \theta_{\text{out}} \Delta \phi_{\text{out}} \Delta \theta_{\text{in}} \Delta \phi_{\text{in}} / \sigma_{\text{scat}}}$. \( \Box \)

Results. By employing singular value decomposition for the backscattering matrix $S_r$, the scattering eigenchannels of the Mie sphere are obtained, of which the first six highest eigenvalues are listed in Table II. Using the same criterion for strongly or weakly scattering channels as above, it is clear that this subwavelength Mie sphere supports no less than $(N_s = 6)$ strongly scattering eigenchannels.

<table>
<thead>
<tr>
<th>eigenchannel (j)</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s_j$</td>
<td>0.673</td>
<td>0.673</td>
<td>0.270</td>
<td>0.256</td>
<td>0.186</td>
<td>0.186</td>
</tr>
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</table>

TABLE II. The eigenvalues of the first six highest scattering eigenchannels for a subwavelength Mie sphere. The normalized diameter of the Mie sphere is $d_s / \lambda = 8 / 9$, and refractive index of the scatterer is 2.

IV. SCATTERING EIGENCHANNELS IN THE NEAR FIELD

To understand these highly scattering eigenchannels hosted by a single nanoparticle, we employ optical WFS to control the light scattering process, by illuminating the scatterer with an structured eigen-wavefront as described by matrix $V$. Here, we focus on the 2D scatterer and the eigenvalues shown in Table II for simplicity. With these structured eigen-wavefronts, we performed full wave simulation to investigate the near-field patterns of these strongly scattering eigenchannels.

FIG. 2. Near-field distribution of the scattering light $|E_{\text{scat}}|$ by exciting different strongly scattering eigenchannels of a single nanoparticle. From the first column to the last column, the frequencies of the incident light are $b/\lambda = 2/7$, $2/5$, $3/5$ and $3/4$, respectively. Note that the value of $|E_{\text{scat}}|$ in each subfigure has been normalized, such that the change of $|E_{\text{scat}}|$ from maximum to minimum is represented by the thermal colors changing from black to white. The results indicate that different resonant leaky modes of the scatterer are excited.

Figure 2 shows the scattering fields inside the nanoparticle for different eigenchannels at four different frequencies $b/\lambda = 2/7$, $2/5$, $3/5$ and $3/4$, respectively. From these results, we see that the excitation of different highly scattering eigenchannels selectively couples light into different resonant leaky modes of the nanoparticle. As the frequency increases, more high-order resonant modes are excited, featured by more complex field patterns inside the scatterer. Therefore, for optical scattering of a single nanoparticle, the existence of more than one highly scattering eigenchannel can be attributed to these resonant leaky modes. This is underlying relationship indicates that optical WFS could be a powerful tool for the con-
trol of light-matter interaction with nanoparticles.
In addition, from the full wave simulation results, we also obtained the ratios of the backscattered power to that of incident power, expressed as $R_j$. The simulation results fully agree with the prediction of the scattering matrix theory, given by

$$R_j = s_j^2,$$  \hspace{1cm} (15)

where $s_j$ denote the eigenvalues listed in Table I.

V. SPECTRAL CORRELATION OF THE SCATTERING EIGENCHANNELS

The spectral property of these strongly scattering channels are related to the temporal response of a nanoparticle. To investigate the temporal response of the scattering light, we again focused on the 2D case for simplicity. Note that, the total scattering power within the detecting range is proportional to

$$P(t) = \langle |E_{\text{far}}(t, \theta_{\text{out}})|^2 \rangle_{\theta_{\text{out}}} \cdot R_{\theta_{\text{tot}}}$$

$$= \text{FFT} \left[ R_{\theta_{\text{tot}}} \int d\omega \langle E_{\text{far}}^*(\omega, \theta_{\text{out}}) E_{\text{far}}(\omega + \Delta\omega, \theta_{\text{out}}) \rangle_{\theta_{\text{out}}} \right],$$  \hspace{1cm} (16)

where $\langle \cdots \rangle_{\theta_{\text{out}}}$ denotes the average over $\theta_{\text{out}}$ within the detected angle range $\theta_{\text{tot}}$. It is seen that the temporal response of the nanoparticle is determined by the spectral correlation of the scattering light, i.e.,

$$C(\omega, \Delta\omega) = R_{\theta_{\text{tot}}} \langle E_{\text{far}}^*(\omega, \theta_{\text{out}}) E_{\text{far}}(\omega + \Delta\omega, \theta_{\text{out}}) \rangle_{\theta_{\text{out}}}. \hspace{1cm} (17)$$

Since the overall scattering light can be decomposed to the scattering eigenchannels (column of $U$), the spectral correlation can be further computed over these eigenchannels. Since different eigenchannels are orthogonal, $C(\omega, \Delta\omega)$ can be derived to be

$$C(\omega, \Delta\omega) = \sum_{i=1}^{N} \alpha_i^*(\omega) \alpha_i(\omega+\Delta\omega) s_i(\omega) s_i(\omega+\Delta\omega), \hspace{1cm} (18)$$

where $\alpha_i(\omega)$ is the coefficient of the incident light coupling into the $i$-th eigenchannel. Clearly, only the strongly scattering channels make significant contributions to the spectral correlation. In other words, the spectral correlation of the highly scattering eigenchannels plays a key role in determining the temporal response of the nanoparticle. We note that similar spectral correlation has been explored for multiply scattered light through randomly scattering media, demonstrating significant differences for different eigenchannels in the strong and weak scattering regimes. [49, 71]. Due to the existence of multiple highly scattering eigenchannels for a single scatterer, the temporal response of a nanoparticle is predictable by investigating the spectral correlation of these eigenchannels.

To investigate the spectral correlation of the eigenchannels, we excite a highly scattering eigenchannels at a specific frequency, and then scan the frequency of incident light to obtain the relative backscattered power $R(\omega)$. From $R(\omega)$, one gets $s_i(\omega) s_i(\omega + \Delta\omega)$ in Eq. 15 to predict the temporal response of a single scatterer, and thus the response to short optical pulses. This is an analogy to time-resolved optical WFS on samples with many nanoparticles, see, e.g., Refs. [72, 73].

Notably, the phase structure on the wavefront is mostly important for optical WFS, and pure-phase control has been applied in many WFS experiments [30, 31, 34, 40, 41, 72, 75]. Following these considerations, we sent purely phase structured light to the scatterer, with the phase structures obtained from the first two strongly scattering eigenchannels at frequency $b/\lambda = 3/5$. Subsequently, we scanned the frequency in the full wave simulation to obtain the scattering light field in the far field. In the end, we calculated to obtain the ratios of the backscattered power to that of incident power $R$ at different wavelengths, and the results for the two eigen-wavefronts are shown in Figs. 3(a) and 3(b), respectively.

![FIG. 3. Relative backscattered power $R$ versus the frequency of the incident light with different eigen-wavefronts. (a) The incident wavefront is loaded with the phase structure of the first eigenchannel ($s_1=0.267$) at frequency $b/\lambda = 3/5$. (b) The incident wavefront is loaded with the phase structure of the second eigenchannel ($s_2=0.216$) at frequency $b/\lambda = 3/5$. The results show that multiple peaks of backscattered power emerge in different spectral ranges relative to the initial frequency $b/\lambda = 3/5$, indicating the coexistence of short range and long range spectral correlations for the highly scattering eigenchannels.](image)
loration exists, which emerges as single peaks, as indicated by the right third and second red arrows in Figs. 3(a) and 3(b), respectively. We have calculated the field patterns at the corresponding peaks of the relative backscattered power, as shown in the insets of Figs. 3(a) and 3(b). From this result, we find that the short range correlation should be related to a specific resonant leaky mode that resides within the limited spectral range, as can be seen from the similarity of the cavity modes by comparing with those at frequency $b/\lambda = 3/5$ shown in Fig. 2. For frequencies far away from the initial frequency $b/\lambda = 3/5$, multiple peaks of relative backscattered power emerge, demonstrating the existence of long range correlation. We have also calculated the field patterns at these long-range peaks of backscattering ratio, as shown in the insets of Figs. 3(a) and 3(b), respectively. From these results, we see that the long range correlations are related to different resonant leaky modes of the scatterer. Besides, by comparing the field patterns of the resonant leaky modes in Figs. 3(a) and 3(b), we see that there is an even number of hot spots horizontally for the cavity modes excited by first eigenchannel in Fig. 3(a), while the number of hot spots is odd horizontally for the cavity modes excited by the second eigenchannel in Fig. 3(b). It indicates that the long range correlation may originate from a series of resonant leaky modes that are of similar spatial features. These intriguing spectral correlations for the highly scattering eigenchannels constitute a first step in novel temporal responses of light scattered by nanoparticles, controlled with optical WFS.

VI. CONCLUSIONS

We have investigated the manipulation of light scattered by single nanoparticles with optical wavefront shaping (WFS). Our results show that wavefront shaping can serve as an efficient knob to turn on different highly scattering eigenchannels intrinsic to the scatterer. These highly scattering channels are found to be related to the different resonant leaky modes of the scatterer, indicating that manipulation of nanoparticle scattering through WFS offers a possible route toward controlling light-matter interaction with individual nanoparticles. Moreover, we have found that both short range and long range spectral correlations exist for the highly scattering eigenchannels of the scatterer, and the correlations are related to different types of resonant leaky modes of the nanoparticle. Our results demonstrate that optical WFS is not only a powerful method for media composed of large numbers of nanoparticles, but also an efficient method to manipulate scattering by a single nanoparticle. Thus, our results offer new perspectives in the understanding of complex scattering processes involving nanoparticles and other resonant optical systems.

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[57] The transition matrix, that originally originates in high-energy physics should not to be confused with the transmission matrix that originates from mesoscopic physics.


