Collective lattice resonances in disordered and quasi-random all-dielectric metasurfaces

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Collective lattice resonances in disordered 2D arrays of spherical Si nanoparticles (NPs) have been thoroughly studied within the framework of the coupled dipole approximation. Three types of defects have been analyzed: positional disorder, size disorder, and quasi-random disorder. We show that the positional disorder strongly suppresses either the electric dipole (ED) or the magnetic dipole (MD) coupling, depending on the axis along which the NPs are shifted. Contrarily, size disorder strongly affects only the MD response, while the ED resonance can be almost intact, depending on the lattice configuration. Finally, random removing of NPs from an ordered 2D lattice reveals a quite surprising result: hybridization of the ED and MD resonances with lattice modes remains observable even in the case of random removing of up to 84% of the NPs from the ordered array. The reported results could be important for rational design and utilization of metasurfaces, solar cells, and other all-dielectric photonic devices.

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1. INTRODUCTION

Strong coupling between lattice modes in arrays of nanoparticles (NPs) and Mie-type oscillations localized within a single NP has attracted significant attention over the last decade. Pioneering theoretical predictions for 1D arrays of Ag NPs [1–3] and consequent experimental verification for 2D arrays of Au NPs [4–6] have given a momentum to a great number of excellent applications of collective lattice resonances in lasers [7–12], biosensors [13–18], emission enhancement [19–22], and color printing [23–26].

To date, most of the studies have considered diffractive coupling between electric dipole (ED) oscillations and Wood–Rayleigh anomalies [27,28] in arrays of classic plasmonic NPs such as Au or Ag. However, quite recently significant attention has been turned to alternative plasmonic materials such as indium-tin-oxide [29], aluminum [30–33], transition metal nitrides [22,34,35], and nickel [36]. The use of these materials makes it possible to tailor a wavelength of collective lattice modes within a wide spectral range, from UV [31] to IR [35], or enable a magneto-optical activity [37,38] that paves the way for a rich variety of novel and promising applications.

In this context, dielectric NPs with both electric and magnetic dipole (MD) resonances [39] represent a case of specific interest. Arrays of all-dielectric NPs have already found a number of excellent applications in photonics and nanotechnology spanning light-guiding [40–42], metamaterials [43], metasurfaces [44–48], mid-infrared filters [49], and others [50]. However, the coupling between localized oscillations in a single dielectric NP and lattice modes has been addressed only quite recently [51–54], with particular attention to ED and MD coupling [55] and overlapping [56] in 2D Si nanodisk arrays.

While various aspects of diffractive behavior of ED and MD resonances in arrays of NPs have been heavily studied in the recent decade [57–60], just a few works have addressed the effects of positional and size disorders [61–63], and only for arrays with pure ED coupling. Quite interesting results have also been reported for lasing [64,65] and solar energy harvesting [62,66–68] in various types of quasi-periodic and aperiodic structures. However, it is a well-known fact that the presence of imperfections in 1D chains of NPs [69,70], 2D structures [71–76], 3D metamaterials [77], and fractal aggregates [78] may lead to various intriguing effects.

In this work, we thoroughly address this problem within the coupled dipole approximation, and study three types of imperfections in 2D arrays of Si nanospheres: (i) disorder in positions of Si nanospheres of the same size; (ii) disorder in sizes of Si
nanospheres arranged in an ordered 2D lattice; and (iii) quasi-ordered 2D arrays of Si nanospheres of the same size. A comprehensive analysis of these scenarios reveals different impact of disorder on ED and MD coupling with lattice modes.

The paper is organized as follows. In Section 2, we provide a theoretical background for the coupled dipole approximation; next, in Section 3 we discuss general features of ED and MD coupling in ordered lattices; then, the impact of positional and size disorder on optical properties of 2D lattices of Si NPs, as well as their quasi-random modifications, are discussed in Section 4; Finally, we draw general conclusions in Section 5.

2. COUPLED DIPOLE APPROXIMATION

Consider an array of $N$ spherical NPs embedded in vacuum. Under the incident plane-wave illumination with electric $E^0$ and magnetic $H^0$ components, the $i$th particle located at $r_i$ acquires electric $d_i$ and magnetic $m_i$ dipole moments that are coupled to other dipoles and to an external electromagnetic field via the coupled dipole equations [39,79,80]:

$$
\mathbf{d}_i = \alpha'_i \left( E^0 + \sum_{j \neq i}^{N} \hat{G}_{ij} \mathbf{d}_j - \sqrt{\mu_0/\varepsilon_0} \sum_{j \neq i}^{N} \hat{G}_{ij} \mathbf{m}_j \right),
$$

$$
\mathbf{m}_i = \alpha''_i \left( H^0 + \sum_{j \neq i}^{N} \hat{G}_{ij} \mathbf{m}_j + \sqrt{\varepsilon_0/\mu_0} \sum_{j \neq i}^{N} \hat{G}_{ij} \mathbf{d}_j \right),
$$

where $\alpha'_i$ and $\alpha''_i$ are ED and MD polarizabilities of the $i$th particle, respectively, $\varepsilon_0$ and $\mu_0$ are the dielectric constant and magnetic permeability of vacuum, $E^0_i = E^0(r_i)$, $H^0_i = H^0(r_i)$, and

$$
\hat{G}_{ij} = A_{ij} \hat{I} + B_{ij} \left( \frac{\mathbf{r}_i \otimes \mathbf{r}_j}{r_{ij}} \right),
\hat{C}_{ij} = D_{ij} \frac{\mathbf{r}_i \times \mathbf{r}_j}{r_{ij}},
$$

where $\hat{I}$ is a $3 \times 3$ unit tensor, $\otimes$ denotes a tensor product, and $A_{ij}$, $B_{ij}$, and $D_{ij}$ are defined as follows:

$$
A_{ij} = \frac{\exp(ikr_{ij})}{r_{ij}} \left( k^2 + \frac{1}{r_{ij}} + \frac{ik}{r_{ij}} \right),
$$

$$
B_{ij} = \frac{\exp(ikr_{ij})}{r_{ij}} \left( -k^2 + \frac{3}{r_{ij}} + \frac{3ik}{r_{ij}} \right),
$$

$$
D_{ij} = \frac{\exp(ikr_{ij})}{r_{ij}} \left( k^2 + \frac{ik}{r_{ij}} \right),
$$

where $r_{ij} = |\mathbf{r}_i - \mathbf{r}_j|$ is the center-to-center distance between the $i$th and $j$th particles, $k = 2\pi/\lambda$ is a wavenumber, and $\lambda$ is a wavelength of external illumination.

ED and MD polarizabilities are explicitly defined as [81]

$$
\alpha'_i = \frac{3i}{2k^3} n \eta \psi_1(nkr_i) \psi'_1(kr_i) - \psi_1(kr_i) \psi'_1(nkr_i),
$$

$$
\alpha''_i = \frac{3i}{2k^3} n \eta \psi_1(nkr_i) \psi'_1(kr_i) - \psi_1(kr_i) \psi'_1(nkr_i),
$$

where $n$ is the refractive index of the NP material, $R_i$ is the radius of the $i$th particle, $\psi_1(x)$ and $\xi_1(x)$ are Riccati–Bessel functions, and prime denotes the derivation with respect to the argument in parentheses.

The electric $d_i$ and magnetic $m_i$ dipoles induced on each NP can be found from the solution of Eq. (1). In this work, we describe the optical response of the arrays of NPs with the extinction efficiency

$$
Q_e = \frac{4k}{I_0 N(R)^2} \text{Im} \sum_{j=1}^{N} \left( d_j \cdot E_j^{0*} + \frac{\mu_0}{\varepsilon_0} m_j \cdot H_j^{0*} \right),
$$

where $I_0$ is the intensity of the incident field, and the asterisk denotes a complex conjugate. Note that in the general case of polydispersity array with $R_i \neq R$, the mean radius $\langle R \rangle = \sum_{i=1}^{N} R_i / N$ is used to define $Q_e$.

The coupled dipole approximation quite accurately describes optical properties of arrays from relatively small Si NPs. Full-wave simulations [82] show that ED and MD are predominant in arrays of Si NPs with $R = 65 \text{ nm}$, and high-order electric and magnetic field oscillations can be ignored in this case, though higher-order multipoles in all-dielectric NPs are pronounced, for example, in large [83–85] and nonspherical [86] single Si NPs, or closely packed arrays of Si NPs [40].

3. ORDERED ARRAYS

We start with the optical properties of a single Si nanosphere. Though it has been widely discussed in the literature, for instance, in Ref. [39], we plot these results for the reader’s convenience. Figure 1(a) shows the refractive index of Si used in calculations [87], while Fig. 1(b) shows the extinction efficiency $Q_e$ for a single Si nanosphere of various radii $R$. For a single sphere, and only in this case, we calculate $Q_e$, taking into account high-order harmonics [81] required for the convergence of the electromagnetic light scattering problem [88]. It can be seen from Fig. 1(b) that indeed, for given sizes, Si nanospheres have distinct and predominant ED and MD resonances in the visible wavelength range. In what follows, we consider arrays from Si nanospheres with $R = 65 \text{ nm}$ radius. However, in the special case of a size disorder, all possible radii of NPs will fall into the range shown in Fig. 1(b), i.e., $50 \text{ nm} \leq R_i \leq 80 \text{ nm}$. Therefore, the coupled dipole approximation can be used with strong confidence.

Next, it is insightful to discuss optical properties of ordered Si nanostructures. Figures 2(a) and 2(b) show two different types of lattices that have been studied in this work: (i) with fixed period along the $x$ axis, $h_x$, and varying period along

![Fig. 1.](image-url)

(a) Refractive index $n$ of Si from Ref. [87]; (b) extinction spectra for a single Si NP of various radii $R$, taking into account high-order multipoles. Spectral positions of ED and MD resonances are denoted as “ed” and “md,” respectively.
and \( \gamma \)ing ED gradually shifts to shorter wavelengths, and the corresponding with lattice modes occurs for MD, while the position of the second case, according to Fig. 2(d), the same strong coupling with resonance slightly shifts to shorter wavelengths for large \( \lambda \) and magnetic \( \alpha \)i anomalies \( \lambda = h_x \) and \( \lambda = h_y \), correspondingly.

4. DISORDERED ARRAYS

A. Types of Disorder

According to Eq. (1), two types of disorder can be distinguished \([69]\): (i) off-diagonal and (ii) diagonal. These types affect either off-diagonal or diagonal elements of the interaction matrix in Eq. (1), respectively. The first type of disorder affects only tensors \( G_{ij} \) and \( C_{ij} \), which are the functions of the NPs’ positions, while the second type of disorder affects only \( \alpha_{i,m} \), which are the functions of NPs’ shape and size.

As was shown for ordered arrays of NPs in Fig. 2, two types of coupling can be distinguished. For fixed illumination, optical response of lattices strongly depends on variations of either \( h_x \) and \( h_y \). Thus, to get more insight, we introduce the off-diagonal disorder in the following manner. We study positional disorder along the \( x \) axis, keeping \( y \) coordinates constant, and vice versa, as shown in Figs. 3(a) and 3(b), respectively. We will refer to these two types of positional disorder as \( x \) disorder and \( y \) disorder, correspondingly. For both cases, we introduce deviations \( \sigma_{xy} \), which characterize a degree of disorder. For each \( i \)th particle with initial \( (x_i, y_i) \) coordinates, we randomly set new coordinates as \( (x_i^{\text{dis}}, y_i) \) for \( x \) disorder and \( (x_i, y_i^{\text{dis}}) \) for \( y \) disorder within the following limits:

\[
x_i - \sigma_x \leq x_i^{\text{dis}} \leq x_i + \sigma_x, \quad \text{and} \quad y_i - \sigma_y \leq y_i^{\text{dis}} \leq y_i + \sigma_y.
\]

Both \( x_i^{\text{dis}} \) and \( y_i^{\text{dis}} \) are randomly generated using a uniform distribution for each \( i \)th NP and for each lattice with given \((h_x, h_y)\). Thus, the effects of positional disorder are uncorrelated.

The schematics of the lattice with diagonal (size) disorder is shown in Fig. 3(c). In this case, we keep original coordinates of each NP, and randomly change the radius \( R_i \) of each \( i \)th NP within the following limits using a uniform distribution:

\[
R_i - \sigma_R \leq R_i^{\text{dis}} \leq R_i + \sigma_R.
\]

Again, as in the case of off-diagonal disorder, \( R_i^{\text{dis}} \) is introduced randomly for each NP and for each lattice configuration, which provides uncorrelated results.

Finally, Fig. 3(d) shows a special combination of diagonal and off-diagonal disorders, which attracts specific interest \([64,65]\). It is a well-known fact that the coupling between a single NP resonance and lattice modes strongly depends on the number of NPs in the array \([89,90]\). However, periodic lattices
of strictly arranged NPs are usually considered to study this finite-sized effect. In our work, we fix the initial coordinates and the sizes of NPs, and randomly remove NPs from the lattice, keeping other NPs untouched. This type of imperfection is somewhat similar to vacancies in crystal structures. In what follows, we will refer to lattices shown in Fig. 3(d) as quasi-random arrays.

We emphasize that each lattice configuration for each type of disorder with given \( \sigma_x, \sigma_y, \) and \( \sigma_R \), or number of NPs removed from the lattice in the case of quasi-random arrays, has been simulated only once, without computing ensemble averages. A reasonable closeness to statistical average has been granted by simulating a large enough number of NPs.

B. Off-Diagonal (Positional) Disorder

Figures 4 and 5 show extinction spectra for arrays of NPs with different degrees of \( x \) and \( y \) disorders. It can be seen that these two types of positional disorder affect the optical properties of NPs in a different way, depending on the coupling regime.

As might be expected from the analysis of Fig. 2(d), the \( x \) disorder significantly affects MD, since the latter strongly couples to the Rayleigh anomaly \( R_A \). Clearly, from Fig. 4, one may observe a slight suppression of the MD with the increasing of the degree of disorder, \( \sigma_x \), both for ED and MD coupling scenarios. It also has to be noted that the coupling of MD and \( R_A \) remains observable even for sufficiently large \( \sigma_x \) in Fig. 4(f), where MD is suppressed. The ED remains almost the same for each case shown in Fig. 4.

Figure 5 shows an expected trend: since ED couples to \( R_A \), the \( y \) disorder affects only the former, keeping MD almost the same for various \( \sigma_y \). However, Figs. 5(e) and 5(f) show almost total suppression of ED for \( \sigma_y = 150 \) nm, while in the case of strong \( x \) disorder shown in Figs. 4(e) and 4(f), MD is quite pronounced.

Finally, Fig. 6 shows a detailed comparison of the extinction spectra for arrays with ED or MD couplings. Indeed, the \( x \) disorder strongly suppresses the MD, while the \( y \) disorder suppresses the ED resonance. Since the ED is generally weaker than the MD, the former almost completely disappears for high degrees of \( y \) disorder. For completeness, Figs. 6(g) and 6(h) show spectra for arrays with \( xy \) disorder, which has been introduced in the same way as the \( x \) and \( y \) disorders, but with simultaneous randomization of both \( x_i \) and \( y_i \) coordinates of each NP. It can be seen that, in general, such a combined disorder yields a superposition of both \( x \) and \( y \) disorders suppresses both ED and MD resonances.

C. Diagonal (Size) Disorder

Figure 7 shows extinction spectra for arrays with various degrees of size disorder, \( \sigma_R \). It is clearly seen that random variations of NP sizes strongly suppress both ED and MD resonances. However, MD remains observable only for \( \sigma_R = 5 \) nm, while for larger \( \sigma_R \), it almost completely disappears. Contrarily, the ED resonance is preserved in all cases, and, of note, EDs strongly couple with Rayleigh anomalies,
Ray, even for high degrees of diagonal disorder, as shown in Fig. 7(e). This effect might be explained by the different behavior of polarizabilities $\alpha_e$ and $\alpha_m$ [39], which yield a different impact of size disorder on ED and MD resonances.

To get a deeper insight, we plot $Q_e$ for arrays with fixed $h_x$ and $h_y$, as shown in Fig. 8. Indeed, Figs. 8(c), 8(e), and 8(g) show that size disorder has a surprisingly weak effect on the ED resonance of arrays with strong ED coupling. It can be seen from Fig. 8(g) that maximum $Q_e$ for the ED resonance drops by no more than 10% for $\sigma_R = 15$ nm compared to the ordered array shown in Fig. 8(a). For arrays with MD coupling, $Q_e$ for ED resonance drops more strongly, by a factor of 2 for $\sigma_R = 15$ nm, as shown in Fig. 8(h). As for the MD resonance, in both the ED and MD coupling cases, the extinction efficiency for MD sharply drops for $\sigma_R = 5$ nm. For larger $\sigma_R$, the MD resonance becomes almost indistinguishable.

**D. Quasi-Random Arrays**

From the previous discussion of diagonal and off-diagonal types of disorder, we can conclude that a simultaneous implementation of positional and size disorders should likely result in the superposition of the effects shown in Figs. 4, 5, and 7. Thus, we do not consider arrays of randomly located NPs of different size. Instead, we introduce a specific combination of positional and size disorders, as shown in Fig. 3(d). These quasi-random arrays are fundamentally different from those shown in Figs. 3(a)–3(c), since random elements of the interaction matrix in Eq. (1) are strictly set to zero in the case of quasi-random arrays, while in previously considered scenarios, off-diagonal or diagonal elements have acquired random deviations according to $\sigma_x$, $\sigma_y$, or $\sigma_R$.

Here, we consider NPs with the same size, $R = 65$ nm, but increase their number to $N = 30 \times 30$ (while previously discussed arrays had $N = 20 \times 20$ NPs). Next, we randomly remove 171, 459, or 756 NPs, leaving the rest 81%, 49%, or 16% of NPs untouched, respectively. We note that the consideration of larger arrays is preferable for this type of disorder, since coupling effects may be totally suppressed in arrays from the small number of NPs left in the lattice [89]. However, in the smallest array considered here, we keep 144 quasi-randomly

![Fig. 6. Extinction spectra $Q_e$ of NPs arrays with ED coupling (left), and MD coupling (right) for various degrees of positional disorder (c) and (d) $\sigma_x$, (e) and (f) $\sigma_y$, and (g) and (h) $\sigma_{xy}$. Corresponding values of $h_x$ and $h_y$ are shown in legends. Dashed vertical lines denote positions of Rayleigh anomalies RA$_y$ at $\lambda = 500$ nm (left), and RA$_x$ at $\lambda = 540$ nm (right).](image)

![Fig. 7. Extinction spectra $Q_e$ for the same 2D lattices as in Figs. 2(c) and 2(d), but for various degrees of size disorder $\sigma_R$, as shown in Fig. 3(c).](image)

![Fig. 8. Extinction spectra $Q_e$ of NP arrays with ED coupling (left), and MD coupling (right) for various degrees of size disorder $\sigma_R$. Corresponding values of $h_x$ and $h_y$ are shown in legends. Dashed vertical lines denote positions of Rayleigh anomalies RA$_y$ at $\lambda = 500$ nm (left), and RA$_x$ at $\lambda = 540$ nm (right).](image)
located NPs, which is sufficient for the emergence of coupling effects.

Intuitively, one could expect the suppression of ED and MD couplings with the increasing number of NPs removed from the ordered array. Indeed, Fig. 9 confirms such an expectation. However, it can be seen that lattices that contain 81% of the initial NPs have almost the same optical properties as the original periodic arrays. Moreover, Figs. 9(e)–9(f) show that ED and MD are coupled to Rayleigh anomalies (though quite weakly) in the arrays with only 16% NPs left, and extinction spectra of such arrays tend to become closer to \(Q_e\) of a single NP.

For comparison, Fig. 10 shows spectra of ordered arrays [as in Figs. 2(a) and 2(b)] from exactly the same number of NPs as in quasi-random arrays, i.e., 27 × 27, 21 × 21, and 12 × 12, and with the same \(h_x\) and \(h_y\). It can be seen from Figs. 10(c) and 10(d) that \(Q_e\) of the quasi-random array from 729 NPs is also almost the same as \(Q_e\) for the periodic 27 × 27 array. Moreover, even with the increasing number of NPs removed from the array, \(Q_e\) of the quasi-random lattices is quite close to that for the strictly ordered arrays with the same number of NPs. However, in the most extreme cases of quasi-random arrays shown in Figs. 10(g) and 10(h), the collective ED resonances are almost suppressed, while the MD coupling remains observable, although the corresponding peak of MD resonance is blue-shifted compared with the ordered arrays.

![Fig. 9. Extinction spectra \(Q_e\) for quasi-random 2D lattices, as shown in Fig. 3(d), for different numbers of NPs. (a) and (b) 81% = 729, (c) and (d) 49% = 441, and (e) and (f) 16% = 144 kept untouched in \(N = 30 \times 30\) arrays of NPs with \(R = 65\) nm. Note the different color scale in the last row, (e) and (f).](image)

![Fig. 10. Extinction spectra \(Q_e\) of NPs arrays with ED coupling (left), and MD coupling (right) for (a) and (b) \(N = 30 \times 30\) array, and for its various quasi-random modifications (solid lines); (c) and (d) 81% = 729, (e) and (f) 49% = 441; and (g) and (h) 16% = 144 (NPs kept untouched). For comparison, \(Q_e\) of strictly periodic (dashed lines) arrays of the same number of NPs are shown; (e) and (d) \(N = 27 \times 27 = 729\); (c) and (f) \(N = 21 \times 21 = 441\); and (g) and (h) \(N = 12 \times 12 = 144\); gray dashed-dotted lines show \(Q_e\) of a single Si NP with \(R = 65\) nm. Corresponding values of \(h_x\) and \(h_y\) are shown in legends. Dashed vertical lines denote positions of Rayleigh anomalies \(RA_x\) at \(\lambda = 500\) nm (left), and \(RA_y\) at \(\lambda = 540\) nm (right).](image)

5. CONCLUSION

We have theoretically analyzed the impact of various types of imperfections on the optical response of 2D arrays of spherical Si NPs. ED and MD resonances are dominant in Si nanospheres in the considered range 50 nm ≤ \(R\) ≤ 80 nm; thus, we have used the coupled dipole approximation, which adequately describes electromagnetic properties of Si NP arrays [82].

We first have shown the existence of two types of collective resonances in 2D arrays emerging from the strong coupling of either ED or MD resonances of a single NP with lattice modes (Rayleigh anomalies) of the 2D array. Such a coupling occurs when the corresponding component of the incident field (electric or magnetic) is orthogonal to the varied period (\(h_x\) or \(h_y\)) of the lattice, while the other period (\(h_y\) or \(h_x\)) is constant [55].

Second, we have shown that electric or magnetic responses are affected by the positional disorder only when NPs are shifted along the axis that is orthogonal to the corresponding component of incident electromagnetic illumination. In our case, for \(\mathbf{E}_0\parallel x\) and \(\mathbf{H}_0\parallel y\), ED and MD resonances are strongly suppressed only for \(y\) or \(x\) disorders, respectively. Obviously, both resonances are affected when NPs are shifted along the \(x\) and \(y\) axes simultaneously.

Next, we have demonstrated that the collective MD response almost completely vanishes in the case of diagonal (size) disorder with \(\sigma_R > 5\) nm. However, the electric counterpart remains quite stable, especially in the case of strong collective coupling between the ED resonance and lattice modes, even for highly polydisperse arrays with \(\sigma_R = 15\) nm.
Finally, we have considered quasi-random arrays as a special combination of off-diagonal and diagonal disorders. Instead of simultaneously shifting the NPs and changing their sizes, we have randomly removed NPs from the lattice, keeping other NPs at original points with original sizes. Surprisingly, arrays with only 16% of NPs left in the lattice exhibit both electric and magnetic collective resonances. However, the extinction spectra of such arrays tend to be similar to spectra of a single NP.

The reported results provide a comprehensive analysis and a fundamental understanding of the impact that disorder has on collective resonances in 2D arrays of all-dielectric NPs. While we have considered spherical Si NPs embedded in vacuum, one could expect similar trends for all-dielectric arrays of NPs of other shapes or materials [91] as long as high-order multipoles can be neglected. Thus, we believe that the reported results may pave the way for future applications in all-dielectric nanophotonics.

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