Hybridization and the origin of the Fano resonances in symmetric nanoparticle trimers

Ben Hopkins,1,∗ Dmitry S. Filonov,2
Stanislav B. Glybovski,2 and Andrey E. Miroshnichenko1,†

1Nonlinear Physics Centre, Australian National University, Acton, ACT 2601, Australia
2ITMO University, St. Petersburg 197101, Russia

Abstract

We study the light scattering by symmetric trimers comprised of either plasmonic or dielectric particles. Such structures have been predicted to produce a pronounced polarization-independent Fano resonance. We provide an alternative approach to hybridization of the trimer’s the doubly-degenerate eigenmodes, and show they exhibit a so-called exceptional point; a topological singularity in their eigenspace. These eigenmodes, subsequently, do not exist independently from each other. By extending our results we are able to confirm that such doubly-degenerate eigenmodes will indeed produce polarization-independent Fano resonances. Experimental measurements of a trimer’s polarization-independent Fano resonance are in good agreement with the expected response from the hybridized eigenmodes and thereby demonstrate the validity of our approach.
I. INTRODUCTION

The Fano resonance\textsuperscript{1} is a relatively-common and well-recognized resonant interference effect in the optical scattering response of many nanoscale structures. Recently, they attracted a lot of attention due to their distinctively-sharp extinction lineshapes, and also due to their peculiar near-field behavior in surface-enhanced Rahman scattering\textsuperscript{2}, nonlinear response\textsuperscript{3}, and enhancement of circular dichroism\textsuperscript{4}. The existence of optical Fano resonances has generally been explained through the use of a hybridization theory argument\textsuperscript{5}, an argument which involves subdividing a structure into two or more subsystems with known properties, and then deducing how their optical responses combine together. The way these optical responses are combined, or hybridized, is regularly depicted as per molecular orbital theory, where the modes of each subsystem are added constructively or destructively to form a bonding and an antibonding mode; the latter of which exhibits suppressed scattering associated with the Fano resonance. However, the applicability of this hybridization approach has only been derived explicitly for systems with high levels of symmetry, such as concentric spheres\textsuperscript{6} and two-particle dimers\textsuperscript{7,8}. It is, nonetheless, a relatively-common practice to utilize the same molecular orbital hybridization when analyzing scattering systems that have lower symmetry. Here we aim to investigate how hybridization translates from simple to more complex geometries. It was recently suggested\textsuperscript{9,10} that polarization-independent Fano resonances should exist in symmetric nanoparticle trimers and, particularly, trimers made of dielectric nanoparticles. Such polarization-independent Fano resonances can also be observed in more generic nanoparticle oligomers\textsuperscript{11}, where the hybridization approach can be performed in equivalent ways for arbitrary polarization angles. The origin of the polarization-independent response in a trimer comes from its discrete rotational symmetry\textsuperscript{12,13}. However, the existence of polarization-independent Fano resonances is much less obvious based on hybridization approach, because it is not possible to subdivide a trimer in a way that conserves its symmetry. In this paper we resolve this by performing the explicit hybridization procedure, analytically, for a symmetric trimer. Our derivations show that the hybridization procedure for a trimer of plasmonic particles can be significantly different to that depicted in molecular orbital theory. However, to investigate the prediction of strong Fano resonances in all-dielectric trimers, we also extend our hybridization approach to include magnetic responses of the individual constituent particles. We then experimen-
tally verify our predictions for the existence of a strong, polarization-independent, Fano resonance in an all-dielectric trimer. Using our approach, we are able to obtain a near-quantitative agreement between the expected response of the hybridized eigenmodes and the experimental measurements. In doing so, we unambiguously demonstrate the validity of our hybridization procedure.

II. THE HYBRIDIZATION OF PLASMONIC NANOPARTICLE TRIMERS

To begin, we take advantage of the fact that a trimer can only be subdivided into a dimer and a single particle; the only other option would be the trivial case of dividing it into three single particles. We will restrict ourselves to the electric dipole responses only of each individual particle, since it dominates for subwavelength plasmonic particles. Under this constraint, the single particle and dimer eigenmodes are shown in Fig. 1a and 1b. We next acknowledge that it is always possible to find an instance of a trimer’s doubly-degenerate eigenmode that is either odd or even under any one of the trimer’s symmetric reflection operations. Now, each of the dimer modes transform according to different irreducible representations of the dimer’s (D$_{2h}$) symmetry group and, as the dimer’s reflection symmetry axis is also shared with that of the trimer, there is a subsequent restriction on which dimer and single particle modes can hybridize with each other. Moreover, to conserve the odd or even response to the dimer’s reflection symmetry operation, the dimer modes $|B_{3u}\rangle$ and $|B_{1g}\rangle$ can only hybridize with $|p_x\rangle$ in the single particle, while $|B_{2u}\rangle$ and $|A_g\rangle$ can only hybridize with $|p_y\rangle$ (refer to Fig. 1). To derive how the modes of the dimer and single particle hybridize we have to consider the radiative coupling between dipoles based on the electric field ($E$) at location $r'$, which is generated by a single electric dipole ($p$) at a location $r$.

$$
E(r') = \frac{k^2}{\epsilon_0} \hat{G}_0(r', r) \cdot p
$$

$$
= \frac{e^{ikR}}{4\pi\epsilon_0 R} \left[ \left( k^2 + \frac{ik}{R} - \frac{1}{R^2} \right) p - \left( k^2 + \frac{3ik}{R} - \frac{3}{R^2} \right) (n \cdot p) n \right]
$$

(1)

Here $k$ is the wavenumber, $n$ is the unit vector pointing from $r$ to $r'$ and $R = |r - r'|$. The relation between a set of dipole moments and the externally-applied electric field can then be modeled accurately using the dipole approximation:

$$
p_i = \alpha_k \epsilon_0 E_0(r_i) + \alpha_k k^2 \sum_{j \neq i} \hat{G}_0(r_i, r_j) \cdot p_j
$$

(2)
FIG. 1. The eigenmodes of (a) a single particle and (b) a dimer with D$_{2h}$ symmetry, when assuming only in-plane electric dipole responses from each particle. The dimer eigenmodes are labeled according to their associated irreducible representation. We also show the location of the extra particle/s needed to form a trimer. In (c) we depict a generalized energy level diagram for these modes and identify the two coupling channels, which have coefficients $A$ and $B$ (see Eq. [3]).

where $\alpha_E$ is the electric dipole polarizability of a particle. This simply states that the total electric field acting on each particle (and hence its dipole moment) is the sum of the externally-applied field and the field radiated by the dipole moments of every other particle. Thus, Eq. [2] allows us to directly calculate the coupling between dimer and single particle modes. Specifically, we are able to calculate the dipole moments induced by the radiated field from the single particle mode and project it onto the dimer modes (which form a complete basis for the dimer’s response), and vice versa. This gives us expressions for the $A$ and $B$ coupling channels shown in Fig.[1].

\[
A = \frac{\alpha_E e^{ikR}}{8\sqrt{2} \pi R} \left[ 3k^2 + \frac{ik}{R} - \frac{1}{R^2} \right] \quad (3a)
\]

\[
B = -\frac{\sqrt{3} \alpha_E e^{ikR}}{8\sqrt{2} \pi R} \left[ k^2 + \frac{3ik}{R} - \frac{3}{R^2} \right] \quad (3b)
\]
Here we have normalized the single particle and dimer modes to make $A$ and $B$ omnidirectional. All other coupling terms between the other eigenmodes of the dimer and single particle are zero, which is due to a geometry-induced symmetry mismatch between basis vectors. It is important to acknowledge that, unlike molecular orbital hybridization, the hybridization of scattering structures does not have a perennial set of basis vectors (i.e., like atomic orbitals) with known symmetry mismatch conditions to identify recurring coupling/hybridization channels in different geometries. Instead, these channels need to be derived for each geometry in order to perform hybridization correctly. We could have, alternatively, started our hybridization procedure from the $|p_y\rangle$ mode in the single particle with $|B_{2u}\rangle$ and $|A_g\rangle$ in the dimer. This would follow the exact same process, but with different $A$ and $B$ coefficients. Here we chose $|p_x\rangle$, $|B_{3u}\rangle$ and $|B_{1g}\rangle$ for completeness, because we will work with $|p_y\rangle$, $|B_{2u}\rangle$ and $|A_g\rangle$ in the next section. We now define an eigenmode, $|\psi\rangle$, of our trimer to be a linear combination of $|p_x\rangle$, $|B_{3u}\rangle$ and $|B_{1g}\rangle$, with complex scalars $a$, $b$ and $c$ as the excitations of each.

\[
|\psi\rangle = a |p_x\rangle + b |B_{3u}\rangle + c |B_{1g}\rangle \tag{4}
\]

When this situation is illustrated (qualitatively) on an energy level diagram, where an eigenmode’s energy level is the the resonant frequency of its eigenvalue, we get Fig. 1c. This depiction of hybridization can be of further use for multipolar analysis because the energy levels of plasmonic dimer modes are known\cite{24} and the single particle’s eigenvalues are also known through the Mie theory\cite{17}. For our case, however, we will continue to restrict ourselves to the low-order electric dipole responses. The dipole moment profile of an eigenmode will, therefore, satisfy the dipole equation (Eq. 2) as

\[
v_i = \alpha_e \varepsilon_0 \lambda v_i + \sum_{j \neq i} \alpha_k k^2 \hat{G}_0(r_i, r_j) \cdot v_j \tag{5}
\]

By substituting Eq. 4 into Eq. 5 we can rewrite Eq. 5 as a system of three linear equations for the coefficients of each dimer and single particle mode. It allows us to use the dimer and single particle eigenmodes as an orthogonal basis for a subset of the trimer’s eigenmodes.

\[
\begin{align*}
  a - Ab - Bc &= \alpha_e \varepsilon_0 \lambda a \\
  \alpha_e \varepsilon_0 \lambda_{B_{3u}} b - Aa &= \alpha_e \varepsilon_0 \lambda b \\
  \alpha_e \varepsilon_0 \lambda_{B_{1g}} c - Ba &= \alpha_e \varepsilon_0 \lambda c
\end{align*}
\tag{6}
\]

\[
\begin{align*}
  (p_x) \\
  (B_{3u}) \\
  (B_{1g})
\end{align*}
\]
where $\lambda_{B_{3u}}$ and $\lambda_{B_{1g}}$ are the eigenvalues of the corresponding eigenmodes in the isolated dimer. These eigenvalues account for self-interaction of the dimer eigenmodes.

$$\lambda_{B_{3u}} = (\alpha E \epsilon_0)^{-1} + \frac{e^{ikR}}{2\pi \epsilon_0 R} \left( \frac{ik}{R} - \frac{1}{R^2} \right)$$

$$\lambda_{B_{1g}} = (\alpha E \epsilon_0)^{-1} + \frac{e^{ikR}}{4\pi \epsilon_0 R} \left( \frac{k^2 + ik}{R} - \frac{1}{R^2} \right)$$

The solutions for $a$, $b$ and $c$ that satisfy the eigenmode equation (Eq. 6) can be calculated directly.

$$|v_1\rangle : \begin{cases} a = \sqrt{2} \\ b = -1 \\ c = \sqrt{3} \\ \lambda_1 = (\alpha E \epsilon_0)^{-1} + \frac{e^{ikR}}{8\pi \epsilon_0 R} \left( 3k^2 + \frac{5ik}{R} - \frac{5}{R^2} \right) \end{cases}$$

$$|v_{2x}, v_{3x}\rangle : \begin{cases} a = \frac{\sqrt{3}e^{ikR}}{4\sqrt{2\pi \epsilon_0 R}} \left( 5k^2 + \frac{3ik}{R} - \frac{3}{R^2} \right) \pm 2\sqrt{6} \delta \\ b = \frac{\sqrt{3}e^{ikR}}{2\pi \epsilon_0 R} \left( k^2 - \frac{3ik}{R} + \frac{3}{R^2} \right) \pm 4\sqrt{3} \delta \\ c = -\frac{3e^{ikR}}{4\pi \epsilon_0 R} \left( k^2 + \frac{3ik}{R} - \frac{3}{R^2} \right) \\ \lambda_2, \lambda_3 = \lambda_0 \mp \delta \end{cases}$$

Here we note that the $|v_2\rangle$ and $|v_3\rangle$ eigenmodes have similar expressions. In this expression, we have defined a central eigenvalue, $\lambda_0$, and a splitting function, $\delta$, that produces the non-degeneracy between the $|v_2\rangle$ and $|v_3\rangle$ eigenmodes.

$$\lambda_0 := (\alpha E \epsilon_0)^{-1} - \frac{e^{ikR}}{16\pi \epsilon_0 R} \left( k^2 - \frac{ik}{R} - \frac{1}{R^2} \right)$$

$$\delta := \frac{1}{\alpha E 16\pi \epsilon_0 R} \left[ 5\alpha^2 E^2 e^{2ikR} \left( \frac{k^2 + \frac{3ik}{R} - \frac{3}{R^2}}{5} \right)^2 + \left( \frac{8k^2}{5} \right)^2 \right]$$

Importantly, if we were to consider these eigenmodes in complex frequency space ($s$-space) to find and/or analyse their resonances\textsuperscript{[13]}, neither $|v_2\rangle$ or $|v_3\rangle$ could be defined for a single sign of $\delta$. Moreover, there are points in $s$-space where the expressions for $|v_2\rangle$ and $|v_3\rangle$ are the same: at $\delta = 0$. At such points, the two eigenmodes coalesce and the eigenspace of these two eigenmodes subsequently reduces in dimension\textsuperscript{[19]}. Such coalescence points are also known as \textit{exceptional points} and the coalescing eigenmodes will interchange expressions with each other depending on the path taken through $s$-space in the vicinity of these
- In our trimer, the locations of four such coalescence points can be found by solving $\delta = 0$ for a complex $k$ in Eq. 10. An investigation into the topology of an eigenspace is well beyond the scope of this work and, instead, it suffices that the eigenmodes and eigenvalues in Eq. 8 are exact expressions for that of a symmetric trimer when considering only the electric dipole response of each individual nanoparticle. We can now categorize these eigenmodes according to their irreducible representation (refer to Table I). Specifically, $|v_1\rangle$ has azimuthally-oriented dipole moments, making it the trimer’s $A_2'$ eigenmode (see Ref. 12), whereas $|v_2\rangle$ and $|v_3\rangle$ are, doubly-degenerate, $E'$ eigenmodes. The two-fold degeneracy of $|v_2\rangle$ and $|v_3\rangle$ can be obtained by either applying a symmetry operation (such as a rotation), or, equivalently; repeating the calculation of trimer eigenmodes from $|B_{2u}\rangle$, $|A_g\rangle$ and $|p_y\rangle$.

- The latter option also has the advantage of providing the, radially-oriented, $A_1'$ eigenmode and its eigenvalue. Therefore, to both justify this statement and provide completeness, we will give the result of repeating the eigenmode calculation to find eigenmodes using, instead, $|B_{2u}\rangle$, $|A_g\rangle$ and $|p_y\rangle$ as the basis vectors.

$$|v\rangle = a' |p_y\rangle + b' |B_{2u}\rangle + c' |A_g\rangle$$ (11)
The coupling channels between these modes are depicted in Fig. 2d, where the $A'$ and $B'$ coupling coefficients can be calculated in the same manner as the $A$ and $B$ coefficients.

\[
A' = \frac{\alpha E e^{i k R}}{8 \sqrt{2} \pi R} \left( k^2 - \frac{5 i k}{R} + \frac{5}{R^2} \right) \quad (12a)
\]

\[
B' = B \quad (12b)
\]

So, by repeating the procedure to find eigenmodes, we obtain the following three eigenmodes.

\[
|v_4\rangle : \begin{cases} 
    a' &= \sqrt{2} \\
    b' &= -1 \\
    c' &= -\sqrt{3} \\
    \lambda_4 &= (\alpha E \epsilon_0)^{-1} - \frac{e^{i k R}}{8 \pi \epsilon_0 R} \left( k^2 + \frac{7 i k}{R} - \frac{7}{R^2} \right)
\end{cases} \quad (13)
\]

\[
|v_{2y}\rangle, |v_{3y}\rangle : \begin{cases}
    a' &= \frac{\sqrt{3} e^{i k R}}{4 \sqrt{2} \pi \epsilon_0 R} \left( k^2 - \frac{9 i k}{R} + \frac{9}{R^2} \right) \pm 2\sqrt{6} \delta \\
    b' &= \frac{3 e^{i k R}}{4 \pi \epsilon_0 R} \pm 4 \sqrt{3} \delta \\
    c' &= -\frac{3 e^{i k R}}{4 \pi \epsilon_0 R} \left( k^2 + \frac{3 i k}{R} - \frac{3}{R^2} \right) \\
    \lambda_2, \lambda_3 &= \lambda_0 \mp \delta
\end{cases} \quad (14)
\]

Here the $|v_4\rangle$ eigenmode belongs to the $A_1'$ irreducible representation and has radially-oriented dipole moments, whereas the $|v_{2y}\rangle$ and $|v_{3y}\rangle$ are, notably, $E'$ eigenmodes that are degenerate with $|v_{2x}\rangle$ and $|v_{3x}\rangle$, respectively. In other words, by using the hybridization approach, we have found two $E'$ eigenmodes for the trimer. This is known to be the maximum number that a ring-type oligomer can exhibit in the dipole approximation\textsuperscript{10}. Therefore we know that the hybridization approach here has determined all of the $E'$ doubly-degenerate eigenmodes of a plasmonic trimer. We can see that their eigenvalues are split by a value of $\delta$ about a central eigenvalue $\lambda_0$, neither of which are eigenvalues of any single mode of the dimer or single particle. Furthermore, using the expressions in Eq. 8 or Eq. 14, one can see that $|v_2\rangle$ and $|v_3\rangle$ are nonorthogonal under complex projections, a result which could be expected given we have a non-Hermitian system. This means that polarization-independent Fano resonant features could indeed be expected to occur in plasmonic trimers, which is quite interesting given neither the dimer or single particle are able to exhibit Fano resonances in isolation (in the dipole approximation). In other words, the coupling channels between dimer and single particle are responsible for the nonorthogonality of the trimer’s $E'$ eigenmodes.
However, this does not complete our analysis because the trimer’s eigenmodes were derived when assuming only electric dipole responses from each particle. Because of this, our analysis can only correspond to plasmonic nanoparticle trimers, whereas Fano resonances have been predicted to be much more substantial in silicon, all-dielectric, trimers. The hybridization approach here is not sufficient for considering all-dielectric oligomers as high-index dielectric nanoparticles produce both electric and magnetic dipole responses. Our theory of trimer hybridization will, therefore, be extended to encompass dielectric materials and the associated magnetic response in the next section.

III. ALL-DIELECTRIC TRIMERS AND MAGNETIC RESPONSE

To account for the magnetic response of individual high-index dielectric nanoparticles we will consider both the electric and magnetic dipole responses of individual particles. For this, we can employ the coupled electric and magnetic dipole approximation. This results in two orthogonal sets of eigenmodes of the trimer; one which will transform under the trimer’s symmetry operations according to the $E'$ irreducible representation and the other which will transform according the $E''$ irreducible representation. The intuitive distinction between these two sets of eigenmodes is that they will be excited by either the electric or magnetic field (for $E'$ and $E''$, respectively) in a normally-incident plane wave. The different irreducible representations also means that the trimer’s symmetry prevents $E'$ eigenmodes coupling to the $E''$ eigenmodes and vice versa. If we could neglect the bianisotropic coupling between electric to magnetic dipoles, the $E'$ eigenmodes would be those that we derived in the previous section and the $E''$ eigenmodes would be analogous to the same eigenmodes, but constructed from magnetic dipoles rather than electric dipoles. However, by using the out-of-plane ($z$) direction, the $z$-oriented magnetic dipoles can be arranged into a doubly-degenerate basis vector that transforms according to the $E'$ irreducible representation and the $z$-oriented electric dipoles can be arranged into a doubly-degenerate basis vector that transforms according to the $E''$ irreducible representation, see Fig. 2. Hence, bianisotropic coupling between electric and magnetic dipoles will allow the electric dipole $E'$ ( $E''$ ) modes to couple with the magnetic dipole $E'$ ( $E''$ ) modes. From this point on, we shall consider only the $E'$ eigenmodes because they are the eigenmodes excited by electric field and can therefore be related to those in the previous section. The procedure for finding the $E''$
eigenmodes, which are excited by the magnetic field, is almost identical upon interchanging the electric and magnetic dipole polarizabilities.

To begin, we extend the hybridization diagram of Fig. 1 to take into account both the new space of $z$-oriented $E'$ responses, and the bianisotropic coupling channels. In Fig. 2a-c, we show the complete set of basis vectors that hybridize to form the $E'$ and $E''$ eigenmodes of the all-dielectric trimer. Then, similar to the previous section, we can divide the basis vectors

**Hybridization for a dielectric trimer**

![Diagram showing hybridization](image)

**FIG. 2.** The eigenmodes of (a) a single particle and (c) a dimer with $D_{2h}$ symmetry, when assuming both electric and magnetic dipole responses from each particle, and neglecting the $z$-direction. The (b) $z$-oriented basis vectors of a trimer, which become doubly-degenerate eigenmodes of the trimer when electric-magnetic coupling is neglected. The dimer and trimer eigenmodes are labelled according to their associated irreducible representation. In (d) we depict a generalized energy level diagram analogous to that in Fig. 1 with coupling channels $A', B', C'$ and $D'$ (see Eqs. 12 and 18) according to their even or odd response under the dimer’s reflection symmetry operation.
However the choice of using the basis vectors that are even or odd is not arbitrary as it was in the previous section. The even (odd) modes offer a convenient opportunity to reduce the number of coupling channels we need to consider for the $E'$ ($E''$) responses. Moreover, as seen in Fig. 2b, the $z$-oriented $E'$ ($E''$) trimer basis vector that is even (odd) under the reflection symmetry, is an eigenmode of the dimer. This dimer eigenmode transforms according to the $B_{2u}$ ($B_{2g}$) irreducible representation and, therefore, it can only couple to $|p_y⟩$ ($|m_y⟩$) mode of the single particle and the other $|B_{2u}⟩$ ($|B_{2g}⟩$) mode of the dimer. We can subsequently draw the energy level and coupling diagram as shown in Fig. 2d. To now find the eigenmodes of a dielectric trimer we can follow a similar procedure to that which we used to find the plasmonic trimer’s eigenmodes from the dimer and single particle eigenmodes. Moreover, we know the magnetic or electric field radiated by an electric or magnetic dipole

\[
H(r') = \frac{1}{\sqrt{\epsilon_0 \mu_0}} e^{ikR} \frac{1}{4\pi R} \left( k^2 + \frac{ik}{R} \right) n \times p
\]

(15)

\[
E(r') = -\frac{\mu_0}{\epsilon_0} \frac{e^{ikR}}{4\pi R} \left( k^2 + \frac{ik}{R} \right) n \times m
\]

(16)

For completeness we will also show the following equation, which is the magnetic field radiated by a magnetic dipole and the partner equation to Eq. 1.

\[
H(r') = \frac{e^{ikR}}{4\pi R} \left[ \left( k^2 + \frac{ik}{R} - \frac{1}{R^2} \right) m - \left( k^2 + \frac{3ik}{R} - \frac{3}{R^2} \right) (n \cdot m)n \right]
\]

(17)

From these we can directly calculate the $C'$ and $D'$ coupling coefficients between each of the dimer and single particle eigenmodes.

\[
C' = \begin{cases} 
\alpha_H \frac{1}{\sqrt{\epsilon_0 \mu_0}} e^{ikR} \frac{1}{4\sqrt{2} \pi R} \left( k^2 + \frac{ik}{R} \right) & \left( C'_1 \right) \\
-\alpha_E \frac{\mu_0}{\epsilon_0} \frac{e^{ikR}}{4\sqrt{2} \pi R} \left( k^2 + \frac{ik}{R} \right) & \left( C'_2 \right)
\end{cases}
\]

(18a)

\[
D' = \sqrt{2} C'
\]

(18b)

We can then define an eigenmode of the dielectric trimer, $|w⟩$, as a linear combination of single particle and dimer eigenmodes.

\[
|w⟩ = a' |p_y⟩ + b' |B_{2u}⟩ + c' |A_g⟩ + d' |E'_z⟩
\]

(19)

Analogous to Eq. 6 for plasmonic trimers, we can then use these eigenmodes as a set of basis
vectors to find a subset of the dielectric trimer’s eigenmodes.

\[
\begin{align*}
\alpha_E \epsilon_0 \lambda_{B_{2u}} b' - A' a' - D'_2 d' &= \alpha_E \epsilon_0 \lambda a' \\
\alpha_E \epsilon_0 \lambda_{A_g} c' - B' a' &= \alpha_E \epsilon_0 \lambda c' \\
\alpha_H \lambda_{E_z} d' - C'_1 a' - D'_1 b' &= \alpha_H \lambda d'
\end{align*}
\]  

(20)

where

\[
\begin{align*}
\lambda_{B_{2u}} &= (\alpha_E \epsilon_0)^{-1} - \frac{e^{ikR}}{4\pi\epsilon_0 R} \left( k^2 + \frac{ik}{R} - \frac{1}{R^2} \right) \\
\lambda_{A_g} &= (\alpha_E \epsilon_0)^{-1} - \frac{e^{ikR}}{2\pi\epsilon_0 R} \left( \frac{ik}{R} - \frac{1}{R^2} \right) \\
\lambda_{E_z} &= \alpha_H^{-1} + \frac{e^{ikR}}{4\pi R} \left( k^2 + \frac{ik}{R} - \frac{1}{R^2} \right)
\end{align*}
\]

The expression for the \( A'_1 \) (\( A''_1 \)) eigenmode, which has radially-oriented electric (magnetic) dipole moments, is straightforward to obtain for this system, simply because it does not couple bianisotropically into anything and is therefore unchanged from the previous section. In other words, \(|v_4\rangle\) remains an eigenmode of the dielectric trimer with eigenvalue \( \lambda_4 \), as per Eq. 13. On the other hand, \(|v_1\rangle\) will not remain an eigenmode as it can couple into a basis vector of uniformly \( z \)-oriented, magnetic dipole moments. This particular coupling between \( A'_2 \) eigenmodes, and its consequences, are investigated separately will be published elsewhere. For our purposes, the important point is that the remaining eigenmodes (\( i.e. \) not belonging to \( A'_1 \)) of Eq. 20 must all transform according to the \( E' \) irreducible representation. This is because the remaining space can be spanned by the two \( E' \) eigenmodes we derived in Eq. 14 and the \(|E'_z\rangle\) basis vector. Any linear combination of these basis vectors must therefore also transform according to the \( E' \) irreducible representation. Indeed, it is follows from Eq. 20 that these three basis vectors are the explicit eigenmodes when \( C'', D' \to 0 \).

On the other hand, outside of this limit, we have to use Eq. 20 to find the expressions of the \( E' \) (and \( E'' \)) eigenmodes. This can be performed using analytical software such as Mathematica, but the result is cumbersome and does not provide intuitive understanding. As such it is easier to use a numerical approach to find eigenmodes of Eq. 20 (considering it as a matrix equation). However, it is still possible to get some conclusion analytically since we know the \( E' \) eigenmodes transform according to a different irreducible representation to that of \(|v_4\rangle\); the \( E' \) eigenmodes must, therefore, satisfy Eq. 20 and be orthogonal to \(|v_4\rangle\). We
can thereby write a general form for the $E'$ eigenmodes:

$$|w_i⟩ : \begin{cases}
    a'_i = a' \\
    b'_i = \sqrt{2}a' - \sqrt{3}c' \\
    c'_i = c' \\
    d'_i = d'
\end{cases} \quad (21)$$

From this we can explicitly see that a dielectric trimer will have three $E'$ eigenmodes, instead of two for plasmonic trimer\(^{30}\). Moreover, a dielectric trimer also has three $E''$ eigenmodes that can be excited by the incident magnetic field. Therefore, the total number of, doubly-degenerate, eigenmodes that can be excited in a dielectric trimer with a normally-incident plane wave is six, which are organized into two sets of three interfering eigenmodes. For comparison, a plasmonic heptamer (a central nanoparticle surrounded by a ring of six nanoparticles) has a single set of three interfering eigenmodes (see Ref.\(^{10}\)). The dielectric trimer geometry therefore has double the propensity for Fano resonances as the plasmonic heptamer geometries. In the next section we will confirm our analysis and demonstrate, experimentally, that a trimer made from high-index dielectric particles is able to exhibit a strong polarization-independent Fano resonance.

IV. EXPERIMENT

The theoretical predictions for the existence of Fano resonances in all-dielectric trimers were discussed for silicon spherical nanoparticles\(^{9,10}\), which exhibited Fano resonances in the optical frequency range. However, it is possible to create a macroscopic analogue of silicon nanospheres in the microwave range, specifically: using MgO-TiO\(_2\) ceramic spheres characterized by dielectric constant of 16 and dielectric loss factor of $(1.12 - 1.17) \times 10^{-4}$ (measured at $9 - 12$ GHz). These ceramic spheres in the microwave range therefore have very similar properties to silicon spheres in the optical range. As such, we are able to create a macroscopic analogue of a silicon nanosphere trimer. This allows us to investigate the scattering properties of a single trimer with much less noise than would be possible for a single silicon nanosphere trimer. In Fig. 3a, we show an experimental setup. The trimer consists of three MgO-TiO\(_2\) spheres with 15mm diameter and 20 mm distance between the centers of the spheres. To fasten together the particles, we use a custom holder made of a styrofoam material with dielectric permittivity of 1 (in the microwave frequency range).
FIG. 3. (a) Experimental setup for the trimer made from MgO-TiO$_2$ spheres and (b) the measurements of extinction for both the trimer and a single sphere. The trimer exhibits a pronounced, polarization-independent, Fano resonance at 4.8GHz in very good agreement with (c) the simulation results of extinction, which were calculated using CST Microwave Studio.

To approximate the plane wave excitation and receive the signal scattered to the forward direction, we employ a pair of, identical, rectangular linearly-polarized wideband horn antennas (TRIM, 1 – 18 GHz), connected to the coaxial ports of a vector network analyzer (Agilent E8362C). The trimer is located in the far-field of both antennas; the distance from the trimer to both the receiving and the transmitting antennas is approximately 1.5 m. The total extinction value is then extracted from the measured complex magnitude of the forward scattered signal by means of the optical theorem. The experimentally measured and numerically simulated (CST Microwave Studio, Time Domain solver) extinction spectra are shown in Fig. 3b and 3c. The extinction spectrum of an isolated MgO-TiO$_2$ sphere was also measured and simulated. In Fig. 3b, one can see a pronounced Fano resonance at 4.8GHz associated with the suppression of the scattering. By varying the orientation of the trimer, it exhibits a polarization independent response. The Fano resonance must subsequently be from interference between doubly-degenerate eigenmodes, and we can, therefore, consider the hybridized eigenmodes from Eq. 20. Moreover, by numerically calculating these eigenmodes we get the extinction spectra shown in Fig. 4a, which accurately reproduces the experi-
ment’s extinction spectrum and Fano resonance. The discrepancy between the hybridization theory and experiment at high frequencies is due to the presence of a known magnetic quadrupole response in the individual spheres, which we simply do not take into account in our approach. Regarding, however, the Fano resonance at 4.8GHz, we are able to decompose the simulated extinction spectrum into components coming from each isolated eigenmode as seen in Fig. 4. The extinction depicted for each eigenmode is ‘isolated’ in the sense that we are neglecting the extinction that can be attributed to the interference between eigenmodes (see Ref. 10). The decomposition clearly shows that there are multiple eigenmodes that interfere destructively to form the main Fano resonance at 4.8GHz. Moreover, modal interference is occurring at very similar frequencies in both the $E'$ and $E''$ response; the Fano feature is coming from interference between $|E'(1)\rangle$ and $|E'(2)\rangle$, and between $|E''(1)\rangle$ and $|E''(2)\rangle$. It is worthwhile to emphasize that this is an overlap of two optical Fano resonances, which are symmetrically-exclusive by nature of their distinct irreducible representations. The likely reason for this situation is that 4.8GHz is also the frequency of an individual sphere’s magnetic dipole resonance, and at least one interfering eigenmode in both the $E'$ and $E''$ responses is dominated by magnetic dipoles. In other words, we have eigenmodes in both irreducible representations that are dependent on a single parameter; the magnetic dipole polarizability of a single MgO-TiO$_2$ sphere. That is to say, the polarizabilities of individual spheres become important when there is non-negligible cross-coupling channels, because it allows them to influence the resonant optical behavior of multiple spaces of optical response. However, more generally, the correlation between a resonance of the single particle and modal interference does appear to be a recurring feature of the experiment. The single particle’s electric resonance at 6.5GHz coincides with destructive interference, albeit only in the $E'$ response between the $|E'(1)\rangle$ and $|E'(3)\rangle$ eigenmodes. We can also see that, in Fig. 3b and 3c, the single sphere’s magnetic quadrupole resonance at 7GHz is also associated with a Fano resonance feature. It is interesting that the eigenmodes in our $E''$ response exhibit, anomalistic, resonant behavior in the vicinity of this, neglected, magnetic quadrupole resonance. Indeed it is very likely that this frequency range should have significant multipolar coupling channels that we have neglected in our hybridization. These observations thereby raise areas of interest for tangent studies into the understanding of multipolar interference and Fano resonances. In any case, the key result for the argument presented in this work is the largest Fano resonance at 4.8GHz, which is fully described by our hybridization theory.
FIG. 4. The (a) extinction of the trimer of Fig. 3 which were calculated using the hybridization procedure from Eq. 20. We also show the eigenmode decomposition of the extinction coming from the $E'$ and $E''$ response of the trimer in (b) and (c). Each eigenmode's isolated contribution to the extinction is shown alongside their associated dipole moment profiles, which depict the real components of each dipole moment at 4.8GHz. Red arrows are electric dipole moments and blue arrows are magnetic dipole moments.

This Fano resonance is a realization of the predicted propensity that all-dielectric trimers have towards Fano resonances.
V. CONCLUSIONS

We have presented an explicit study on the hybridization of optical responses in both plasmonic and all-dielectric trimers. The hybridization procedure we followed for trimers is substantially different from high-symmetry systems, such as concentric spheres and dimers. A general difference is that a hybridized eigenmode necessarily needed to be constructed from multiple eigenmodes from each subsystem. Furthermore, the specific linear combination of each subsystem’s eigenmodes, which collectively formed a hybridized eigenmode of the overall trimer, is neither obvious nor intuitive. However, the most crucial difference is related to the doubly-degenerate $E'$ eigenmodes. The central energy level, about which the eigenvalues of the $E'$ eigenmodes are split, do not correspond to any single eigenvalue of a subsystem. Moreover, the expressions for the trimer’s hybridized eigenmodes could not generally be labelled as either bonding or antibonding, because the expressions are interchangeable with each other depending on the path taken in the vicinity of a coalescence point in their eigenspace. What we have presented here subsequently demonstrates that the conclusions from a molecular orbital hybridization procedure do not to correctly predict the complete properties of a trimer’s doubly-degenerate eigenmodes. By then demonstrating a key prediction of our hybridization theory in an experiment - that a trimer can exhibit a polarization-independent Fano resonance; we are able to verify our hybridization approach. Our conclusions subsequently raise a point of contention against the use of molecular orbital hybridization procedures to deduce the eigenmodes of nonsymmetric systems; a more considered utilization of hybridization concepts appears to be necessary.

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\* ben.hopkins@anu.edu.au
\+ andrey.miroshnichenko@anu.edu.au

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30 In previous work, specifically Ref.10, the increased number of eigenmodes in all-dielectric oligomers due to bianisotropic coupling into \( z \)-oriented dipoles was neglected. In that work the coupled electric and magnetic dipole equations were separated into distinct electric and magnetic equations that have bianisotropic driving terms. Using this depiction, it was shown that there are only two pairs of doubly-degenerate \( E' \) or \( E'' \) eigenmodes that can affect the extinction cross-section. However, as was explained here, that same space must comprise three
(or more) electromagnetic eigenmodes in the presence of non-negligible bianisotropic coupling. Therefore, when taking into account that there will be interference between the electromagnetic eigenmodes, the approach in the aforementioned work will not be able to correctly distinguish modal interference from regular excitation. In addition to the desire for a more fundamental picture, the need to distinguish these can be of significance for observable effects such as the circular dichroism (e.g. see Ref.3).

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