I. INTRODUCTION

Chirality describes the structural properties of three-dimensional objects that cannot be superimposed onto their mirror images [1]. It has been proved that an infinite number of pseudoscalars can be constructed to quantify the chirality of a geometric object [2]. With just one nonzero pseudoscalar, the object is chiral. Therefore, no golden standard is available to quantitatively evaluate or compare the degree of geometric chirality. Instead of geometry, an alternative way to evaluate chirality of an object is based on its light-matter interaction. For example, a chiral object may respond differently to left- and right-handed CPL. This technique has been widely used to characterize the chirality of chemical and biomolecules whose CD signals are mostly very weak [3]. In this work, we develop a multiscattering model that can realize the upper limit of the achievable scattering CD.

On the other hand, a more general definition of CD has been proposed to describe the chiral response of objects under arbitrary light fields rather than the two orthogonal CPL states [14]. Using this general parameter, the maximum CD can also be realized by using light field engineered to match the chiral features of a given object instead of designing structural objects. Optical chirality (also called Lipkin’s 00-zilch [15]) is defined as $C = (\varepsilon_0 E \cdot V \times E + \mu_0 H \cdot V \times H)/2$, describing the local chiral feature of a light field [14]. For a point chiral dipole, the intensity of the CD signal is proved to be proportional to the local $C$ value. Thus, in the past decade a significant amount of effort has been devoted to realize superchiral light with larger $C$ values than CPL beams to enhance CD signals [14,16–20]. In Ref. [7], the chiral response of nanospheres was analyzed. The theoretical upper limit of CD is $g = 2$ (e.g., when $W^-$ is zero but $W^+$ is nonzero), indicating that the object can interact with one handedness of CPL beam, but not with the opposite handedness [12]. For instance, a complex metasurface was designed theoretically to obtain the maximum CD enabled by bound states in the continuum [13]. This maximum CD effect can be interpreted as the perfect match between the geometrical chirality of the designed structures and the CPL field.
CD signal for a Mie sphere made of chiral materials. Under the designed optimized optical fields, the upper limit of CD value (i.e., $g = 2$) can be realized at specific frequencies due to the excitation of anapole states.

II. MULTISCATTERING MODEL FOR CHIRAL SPHERE

To predict the scattering CD from Mie spheres, here we propose a multiscattering model based on the Lorentz-Mie theory [23]. When the chirality of a sphere is weak, the influence of its chiral property can be considered as a perturbation in the model [8]. The basic route of this method contains two steps: Step 1 is to analyze the reference structure by removing the chirality of the sphere [Fig. 1(a)]. The zeroth-order scattering field and the chiral features to solve the scattering CD by sphere with weak chirality when the incident field consists of VSHs with the order of (1, 1).

In Fig. 1(b), the secondary scattering fields by two spheres with opposite chiralities (i.e., enantiomers) are compared. By switching the chirality of the sphere, the sign of scattering field is changed for odd-order terms, while unchanged for even-order terms. Using this multiscattering model, the total scattering field can be decomposed into the chiral part and nonchiral part rigorously. Next, the details of this model are discussed.

The constitutive relations of the reciprocal and chiral media inside a homogeneous sphere can be expressed as [24]

$$ D = \varepsilon_s E - ik_s H, $$

$$ B = \mu_s H + ik_s E. $$

Here $\varepsilon_s$ and $\mu_s$ are the permittivity and permeability of sphere; $k_s$ is the chiral parameter. The scattering field by removing the chiral feature of the sphere (i.e., $k_s = 0$) is considered first. In the frame of Lorentz-Mie theory [23], the scattering field by a Mie sphere should be expanded as the linear combination of the vector spherical harmonics (VSHs) as

$$ E_{\text{scat}}^{(0)} = \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ d_{mn}^{(0)} M_{mn}(k_0, \mathbf{r}) + b_{mn}^{(0)} N_{mn}(k_0, \mathbf{r}) \right], $$

$$ H_{\text{scat}}^{(0)} = \frac{1}{iZ_0} \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ d_{mn}^{(0)} M_{mn}(k_0, \mathbf{r}) + b_{mn}^{(0)} N_{mn}(k_0, \mathbf{r}) \right]. $$

In Eqs. (3) and (4), $k_0$ and $Z_0$ are the vacuum wave vector and vacuum impedance. $d_{mn}^{(0)}$ and $b_{mn}^{(0)}$ are the scattering coefficients. The expressions with the superscript of "(0)" are associated with the reference structure, i.e., the nonchiral sphere in free space. The incident field and the internal field of the sphere can also be expressed as the combination of VSHs, i.e.,

$$ E_{\text{inc}} = \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ u_{mn}^{(0)} R_{g} M_{mn}(k_0, \mathbf{r}) + v_{mn}^{(0)} R_{g} N_{mn}(k_0, \mathbf{r}) \right], $$

$$ H_{\text{inc}} = \frac{1}{iZ_0} \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ u_{mn}^{(0)} R_{g} N_{mn}(k_0, \mathbf{r}) + v_{mn}^{(0)} R_{g} M_{mn}(k_0, \mathbf{r}) \right], $$

$$ E_1 = \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ c_{mn}^{(0)} R_{g} M_{mn}(k_1, \mathbf{r}) + d_{mn}^{(0)} R_{g} N_{mn}(k_1, \mathbf{r}) \right], $$

$$ H_1 = \frac{1}{iZ_1} \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ c_{mn}^{(0)} R_{g} N_{mn}(k_1, \mathbf{r}) + d_{mn}^{(0)} R_{g} M_{mn}(k_1, \mathbf{r}) \right]. $$

To avoid the singularity at the origin (i.e., the center of the sphere), the incident field and internal field are expressed as the combination of regular VSHs (i.e., $R_{g} M_{mn}$, $R_{g} N_{mn}$). In Eqs. (7) and (8), $k_1 = (\varepsilon_s/\mu_s)^{1/2} k_0$ and $Z_1 = (\mu_s/\varepsilon_s)^{1/2}$. By
applying the boundary conditions at the surface of the sphere, the unknown expansion coefficients in scattering field and the internal field can be calculated by $u_{mn}^{(0)}$ and $v_{mn}^{(0)}$:

$$d_{mn}^{(0)} = T_n^a u_{mn}^{(0)},$$  

(9)

$$b_{mn}^{(0)} = T_n^b v_{mn}^{(0)},$$  

(10)

$$c_{mn}^{(0)} = T_n^c v_{mn}^{(0)},$$  

(11)

$$d_{mn}^{(0)} = T_n^d v_{mn}^{(0)},$$  

(12)

In Eqs. (9)–(12), the coefficients of $T_n^a$, $T_n^b$, $T_n^c$ and $T_n^d$ are given in Appendix A. When the chiral property is introduced into the sphere, the scattering field will deviate from the nonchiral case (i.e., in the reference structure). In our model, the terms including $\kappa$, in Eqs. (1) and (2) are considered as the perturbing sources that produce the localized distributions of current $J^{(1)}$ and magnetization $M^{(1)}$ inside the sphere. These perturbing sources are expressed as $J^{(1)} = -i\omega k_1 H_1$ and $M^{(1)} = i(\kappa_1 / \mu) E_1$. It should be noted that $[E_1, H_1]$ are calculated in the sphere with chirality. For materials with weak chirality (i.e., a usual case for existing natural materials), the internal field of $[E_1, H_1]$ can be approximately replaced by $[E_0^{(1)}, H_0^{(1)}]$ in Eqs. (7) and (8). The radiation field of $[E_{inc}^{(1)}, H_{inc}^{(1)}]$ by $J^{(1)}$ and $M^{(1)}$ can be obtained by solving the following inhomogeneous wave equations:

$$(\nabla^2 + k_1^2) \left[ \mathbf{r} \cdot E_{inc}^{(1)} \right] = k_1 Z_1 \mathbf{L} \cdot \left[ M^{(1)} + \frac{1}{k_1^2} \mathbf{V} \times J^{(1)} \right],$$  

(13)

$$(\nabla^2 + k_1^2) \left[ \mathbf{r} \cdot H_{inc}^{(1)} \right] = -i \mathbf{L} \cdot \left[ J^{(1)} + \mathbf{V} \times M^{(1)} \right].$$  

(14)

In Eqs. (13) and (14), the angular momentum operator is defined as $\mathbf{L} = -i \mathbf{r} \times \nabla$. The Green’s function for the wave equations is $G_1(\mathbf{r}, \mathbf{r'}) = e^{i k_1 |\mathbf{r}-\mathbf{r'}|/|\mathbf{r}-\mathbf{r'}|}$, which can be expanded by spherical harmonics as [25]

$$G(\mathbf{r}, \mathbf{r'}) = i k_1 \sum_{n=0}^{+\infty} \sum_{m=-n}^{n} j_n(k_1 r') h_n^{(1)}(k_1 r) \sum_{m=-n}^{n} (-1)^m (2n+1) Y_n^m(\theta', \phi') Y_n^m(\theta, \phi),$$  

(15)

which is only applicable for $r > r'$. In this work, the scalar spherical harmonics are defined as $Y_n^m = P_n^m(\cos \theta) e^{i m \phi}$. At the inner surface of the sphere (i.e., $|\mathbf{r}| = R_1$), the secondary incident field radiated by $J^{(1)}$ and $M^{(1)}$ is expressed as the sum of outgoing VSHs:

$$E_{inc}^{(1)} = \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ u_{nm}^{(1)} M_{mn}(k_1, \mathbf{r}) + v_{mn}^{(1)} N_{mn}(k_1, \mathbf{r}) \right],$$  

(16)

$$H_{inc}^{(1)} = \frac{1}{i Z_1} \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ u_{nm}^{(1)} N_{mn}(k_1, \mathbf{r}) + v_{mn}^{(1)} M_{mn}(k_1, \mathbf{r}) \right].$$  

(17)

Following the derivation in the textbook [25], the expansion coefficients can be calculated by the integrals over the secondary radiation sources:

$$u_{mn}^{(1)} = \frac{k_1^2 Z_1}{Y_{mn}} \int V Y_n^m(\theta, \phi) \left\{ j_n(k_1 r) \left[ (\mathbf{r} \times J^{(1)}) \right] + \frac{1}{r} \left[ r j_n(k_1 r) \right] (\mathbf{r} \cdot M^{(1)}) \right\} d^3 \mathbf{r},$$  

(18)

$$v_{mn}^{(1)} = \frac{i k_1^2 Z_1}{Y_{mn}} \int V Y_n^m(\theta, \phi) \left\{ \frac{1}{r} \left[ r j_n(k_1 r) \right] \left[ \mathbf{r} \times J^{(1)} \right] + i k_1 j_n(k_1 r) (\mathbf{r} \times M^{(1)}) \right\} d^3 \mathbf{r}.$$  

(19)

After some mathematical derivations, Eqs. (18) and (19) can be simplified as

$$u_{mn}^{(1)} = \kappa_1 F_n(k_1 R_1) d_{mn}^{(0)},$$  

(20)

$$v_{mn}^{(1)} = \kappa_1 F_n(k_1 R_1) e_{mn}^{(0)}.$$  

(21)

The definition of $F_n(x)$ is $F_n(x) = -i (\epsilon_1 \mu_1)^{-1/2} [\psi_n^*(x) \psi_n(x) + 2 \int_0^x \psi_n^2(\alpha) d\alpha]$. The secondary incident field radiates from the internal sphere region. The scattering process by sphere surface should be considered in our model. The transmission and reflection field are defined as scattering field and internal field excited by the first-order perturbing sources. These fields are indicated by the superscript “(1).” These first-order perturbing fields can also be expanded by VSHs as

$$E_{scat}^{(1)} = \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ a_{mn}^{(1)} M_{mn}(k_0, \mathbf{r}) + b_{mn}^{(1)} N_{mn}(k_0, \mathbf{r}) \right],$$  

(22)

$$H_{scat}^{(1)} = \frac{1}{i Z_0} \sum_{n=1}^{+\infty} \sum_{m=-n}^{n} \left[ a_{mn}^{(1)} N_{mn}(k_0, \mathbf{r}) + b_{mn}^{(1)} M_{mn}(k_0, \mathbf{r}) \right].$$  

(23)
The expansion coefficients can be calculated to be

\begin{align}
    a_m^{(1)} &= s_n^{(1)} u_m^{(1)}, \\
    b_m^{(1)} &= t_n^{(1)} v_m^{(1)}, \\
    c_m^{(1)} &= s_n^{(1)} t_m^{(1)}, \\
    d_m^{(1)} &= t_n^{(1)} v_m^{(1)}. 
\end{align}

In Eqs. (26–29), the coefficients of $S_n^{(1)}$, $S_n^{(2)}$, $S_n^{(3)}$, and $S_n^{(4)}$ are given in Appendix A. As shown in Fig. 1(b), when the chirality of the sphere is inverted (i.e., $\kappa_s \rightarrow -\kappa_s$), the sign of scattering field $[E_1^{(1)}(r), H_1^{(1)}(r)]$ will be changed. The scattering circular dichroism is introduced by the secondary scattering field. Meanwhile, the internal field $[E_1^{(1)}(r), H_1^{(1)}(r)]$ can produce second order current $J^{(2)}$ and magnetization $M^{(2)}$, whose magnitude is related to $\kappa_s^2$. Therefore, the scattering field $[E_1^{(2)}(r), H_1^{(2)}(r)]$ produced by $J^{(2)}$ and $M^{(2)}$ are nonchiral. Following this procedure, the high order sources can also be calculated. The total scattering field can be expressed as the sum of all the terms associated with different orders of perturbing sources. The total scattering coefficients $a_m$ and $b_m$ can be divided into nonchiral and chiral parts, respectively, i.e.,

\begin{align}
    a_m(\pm \kappa_1) &= A_m \pm A'_m, \\
    b_m(\pm \kappa_1) &= B_m + B'_m, 
\end{align}

where $m$ and $n$ are integers to denote the orders of VSH field. Here $A_m$ and $B_m$ are nonchiral parts, while $A'_m$ and $B'_m$ are chiral parts. The nonchiral parts of $a_m$ and $b_m$ are composed by the even-order coefficients, which are unchanged during the chirality inversion, while the chiral parts are contributed by the odd-order coefficients.

\begin{align}
    A_m &= \Gamma_n^{(a)} u_m, \\
    A'_m &= \gamma_n^{(a)} v_m, \\
    B_m &= \Gamma_n^{(b)} v_m, \\
    B'_m &= \gamma_n^{(b)} u_m. 
\end{align}

In Eqs. (32–35), the coefficients of $\Gamma_n^{(a)}$, $\gamma_n^{(a)}$, $\Gamma_n^{(b)}$, and $\gamma_n^{(b)}$ can be expressed as

\begin{align}
    \Gamma_n^{(a)} &= \left[ T_n^{(a)} + \frac{\kappa_1^2 F_n^{(a)} s_n^{(a)} T_n^{(a)}}{1 - \kappa_1^2 F_n^{(a)} s_n^{(a)} T_n^{(a)}} \right], \\
    \gamma_n^{(a)} &= \frac{\kappa_1 F_n^{(a)} s_n^{(a)} T_n^{(a)}}{1 - \kappa_1^2 F_n^{(a)} s_n^{(a)} T_n^{(a)}}. 
\end{align}

The total scattering power by the chiral sphere can be calculated by the scattering coefficients of $a_m$ and $b_m$ in Eq. (40) [23].

\begin{align}
    W = \frac{1}{2k_0^2} \sum_{m=1}^{\infty} \sum_{n=-\infty}^{\infty} |a_m|^2 + |b_m|^2. 
\end{align}

As discussed in the Introduction, the generalized CD of a chiral sphere can be determined by values of $W^+$ and $W^-$, which are the scattering power under the illuminations of arbitrary field and the corresponding mirror field. By taking the mirror operation on the chiral sphere and optical field simultaneously, the scattering power remains the same. Therefore, $W^+$ and $W^-$ are equal the scattering powers of the chiral sphere with $\kappa_s$ and the mirror sphere with $-\kappa_s$ under the same incident field in Eq. (41),

\begin{align}
    W^\pm = \frac{1}{2k_0^2} \sum_{m=1}^{\infty} \sum_{n=-\infty}^{\infty} |A_m \pm A'_m|^2 + |B_m \pm B'_m|^2. 
\end{align}

Therefore, based on Eqs. (32–35) in the multiscattering model, the value of the scattering CD can be directly calculated by the expansion coefficients of $u_m$ and $v_m$.

III. PREDICTION OF MAXIMUM SCATTERING CD

Next, we implement this model to predict the maximum CD scattering condition for a homogeneous chiral sphere. When the incident field is a linear combination of two regular VSHs with the same order, i.e., $u_m R G M_{mn} + v_m R G N_{mn}$, the scattering CD can then be expressed as

\begin{align}
    g_{mn} = \frac{4 \text{Re} \left[ (\Gamma_n^{(a)} y_n^{(a)} + \Gamma_n^{(b)} y_n^{(b)}) u_m v_m^* \right]}{\left( |\Gamma_n^{(a)}|^2 + |\gamma_n^{(a)}|^2 \right) |u_m|^2 + \left( |\Gamma_n^{(b)}|^2 + |\gamma_n^{(b)}|^2 \right) |v_m|^2}. 
\end{align}

Using Eq. (42), one can obtain the upper limit of the CD value related to the order of $(m, n)$. It can be proved that the upper limit of $|g_{mn}|$ should be $G_n$, which can be expressed as

\begin{align}
    G_n = \frac{2 |\Gamma_n^{(a)} y_n^{(a)} + \Gamma_n^{(b)} y_n^{(b)}|}{\sqrt{ |\Gamma_n^{(a)}|^2 + |\gamma_n^{(a)}|^2 \sqrt{ |\Gamma_n^{(b)}|^2 + |\gamma_n^{(b)}|^2 \right.}}, 
\end{align}

The condition to obtain the maximum value of $g_{mn} = G_n$ is

\begin{align}
    \frac{u_m}{v_m} = e^{-i \phi} \sqrt{ |y_n^{(a)}|^2 + |\gamma_n^{(a)}|^2 \sqrt{ |y_n^{(b)}|^2 + |\gamma_n^{(b)}|^2 \right.}}, 
\end{align}

where $\phi$ is the interference phase.
where $\Psi$ is the complex phase of $\Gamma_n^{(a)} r_n^{(a)*} + \Gamma_n^{(b)} r_n^{(b)*}$. Using Eq. (43), the upper limit of scattering CD, $G_1$, for the order of $n = 1$, is plotted in Fig. 1(c). In this calculation, the radius of the sphere is 320 nm, $\epsilon_1 = 3, \mu_1 = 1$, and $\kappa_1 = 0.01$. Intriguingly, one can see two peaks at the frequencies of 241.46 and 294.46 THz, where the CD values can reach the maximum value of $g = 2$. Importantly, using Eq. (44), one can determine the incident field for the Mie sphere to realize these maximum CD scattering values, as will be explained next.

To determine the required optical field to realize the maximum scattering CD, here we calculate the scattering CD spectra. The incident field consists of two VSHs with ($\Gamma_n^{(a)} r_n^{(a)*} + \Gamma_n^{(b)} r_n^{(b)*}$). Using Eq. (44), the peak condition for $G_1 = 2$ in Fig. 1(c) were realized when $u_{inc}/v_{inc} = 1/55$ and $u_{inc}/v_{inc} = -26$. The scattering spectra of the chiral sphere under $\mathbf{E}^{(+)1}$ are shown by the red curves in Figs. 2(a) and 2(b), respectively. To obtain the CD value, the scattering spectra should be calculated under the incident field after mirror operation, i.e., $\mathbf{E}^{(-)1} = u_{inc} R_g \mathbf{M}_{-1,1} + v_{inc} R_g \mathbf{N}_{-1,1}$. Under the illumination of $\mathbf{E}^{(-)1}$, the scattering spectra of the chiral sphere are shown by the blue curves in Figs. 2(a) and 2(b). In these cases, the scattering power drops to zero at the frequencies of 241.46 and 294.46 THz, indicating the completely suppressed scattering field from the sphere. The corresponding scattering CD spectra for these two pairs of incident fields are calculated using $g = 2(W^+ + W^-)/(W^+ + W^-)$ as shown in Figs. 2(c) and 2(d). One can see that the maximum scattering CD signal is achieved in the sphere enabled by the complete nonscattering states. In contrast, under the illumination of CPL, the value of scattering CD from the same sphere is $-0.114-0.054$ in the frequency range [176.5, 333.5 THz] (see Appendix C).

It should be noted that although the upper limit of scattering CD in Eq. (43) is only applicable for the incident field of $u_{inc} R_g \mathbf{M}_{mn} + v_{inc} R_g \mathbf{N}_{mn}$ with a given order of $(m, n)$, it can be generalized to cases with arbitrary incident fields of $\sum_{n=1}^{N} \sum_{m=-n}^{n} u_{inc} R_g \mathbf{M}_{mn} + v_{inc} R_g \mathbf{N}_{mn}$. For simplicity, the incident field is only consisting of two orders of VSHs. Therefore, the scattering powers under two incident fields (i.e., mirror images of each other) can be expressed as

$$W^+ = W_1^+ + W_2^+,$$

$$W^- = W_1^- + W_2^-.$$

The following two parameters are defined as

$$g_1 = 2(W_1^+ - W_1^-)/(W_1^+ + W_1^-),$$

$$g_2 = 2(W_2^+ - W_2^-)/(W_2^+ + W_2^-).$$

Because the scattering power is positive, $g_1 \in [-2, 2]$ and $g_2 \in [-2, 2]$. The scattering CD can be expressed as

$$g = g_1(W_1^+ + W_1^-) + g_2(W_2^+ + W_2^-)$$

$$W = W_1^+ + W_1^- + W_2^+ + W_2^-$$

Without loss of generality, we assume that $g_1 > g_2$, then

$$g = g_1 + g_2(W_2^+ + W_2^-) \leq g_1,$$

$$g = g_2 + g_1(W_1^+ + W_1^-) \leq g_1.$$

Therefore, it can be proved that $\min(g_1, g_2) \leq g \leq \max(g_1, g_2)$. This conclusion can easily be generalized to the case with more orders of VSHs, i.e., $\min(g_1, g_2, \ldots, g_n) \leq g \leq \max(g_1, g_2, \ldots, g_n)$. Therefore, the absolute value of $g_{mn}$ is limited by $G_n$, so that $\min(-G_1, -G_2, \ldots, g) \leq g \leq \max(G_1, G_2, \ldots, g)$.

IV. CHIRALITY-SENSITIVE ANAPOLE

To reveal the physics of the maximum CD under the designed optical field, we then employ the multipole expansion method to model the chiral scattering [21,25]. The scattering field is radiated by the oscillation current $\mathbf{J}$ and magnetization $\mathbf{M}$ which are excited by the field inside the sphere. The scattering fields are mainly radiated by the magnetic dipole (MD, induced by $\mathbf{J}$), electric dipole (ED), and toroidal dipole (TD). For the maximum scattering CD condition with $u_{inc}/v_{inc} = -26$, amplitudes of the three moments for the sphere as well as the electric and magnetic scattering coefficients (i.e., $a_1$ and $b_1$) under the incident field of $\mathbf{E}^{(-)1}$ are calculated in Fig. 3. At the frequency of 294.46 THz, both scattering coefficients $a_1$ [Fig. 3(a)] and $b_1$ [Fig. 3(b)] are zero, indicating that the sphere is at the completely nonradiating state. Specifically, $a_1$ can be well predicted by the amplitude of MD [Fig. 3(a)], while $b_1$ is mainly contributed by ED and TD [see the amplitudes of $b_1$, ED, and TD in Fig. 3(b)], and phases of ED and TD in Fig. 3(c)]. Specifically, at $f = 294.46$ THz, the amplitudes of ED and TD are equal, but their phases are the opposite [see the vertical dashed line in Fig. 3(c)]. As a result, identical radiation patterns of ED and TD in the far field can
lead to the completely destructive interference, resulting in the excitation of anapole states [26–32]. With the illumination of $E^{(-)}$, the amplitude of the MD is almost unchanged as shown in Fig. 3(d). However, the anapole state is broken because the components of the scattering field contributed by ED and TD cannot be canceled mutually in the far field under the illumination of $E^{(+)}$ (i.e., $b_1 = B_1 - B'_1$). Therefore, the nonradiating state is due to the destructive interference not only between the scattering field components radiated by ED and TD, but also between the chiral and nonchiral parts of the scattering field. In contrast, if the incident field is switched to $E^{(+)}$, the electric scattering coefficient becomes nonzero (i.e., $b_1 = B_1 + B'_1$). In this situation, the scattering fields radiated by the ED and TD cannot be canceled mutually in the far field under the illumination of $E^{(+)}$. As a result, the condition of the anapole state is not fulfilled as shown in Figs. 3(e) and 3(f).

For the other peak condition at $f = 241.46$ THz with $v_{\text{inc}}/u_{\text{inc}} = 1/55$, the mechanism of maximum scattering CD can also be explained by the excitation of anapole states using the multipole expansion method (see Appendix D). It should be noted that the actual conditions for these two peaks at $u_{\text{inc}}/v_{\text{inc}} = -26$ and 1/55 are different; i.e., when the chirality of the incident field is inverted, the anapole state can be maintained for the peak at $u_{\text{inc}}/v_{\text{inc}} = 1/55$ but not for $u_{\text{inc}}/v_{\text{inc}} = -26$. Nevertheless, this subtle difference in physics does not change the key mechanism of the maximum scattering CD, i.e., the excitation of anapole states. Next, we employ numerical simulation to demonstrate the feasibility to realize the maximized CD response of the Mie chiral sphere on substrate. The influence of the chiral parameter on the excitation condition of maximized CD will also be analyzed.

V. NUMERICAL SIMULATION RESULTS

In this section, the numerical simulation based on $T$-matrix method is used to verify the maximized scattering CD of the Mie chiral sphere on substrate in Fig. 4(a) [33,34]. The parameters of the sphere (i.e., $R$, $\varepsilon_r$, $\mu_r$, and $\gamma$) remain the same as in Sec. III. The refractive index of the substrate is...
plane of the incident light is selected at 294.46 THz, which has been 1.37. In Fig. 4(b), the scattering CD is calculated when the ratio of $v_{inc}/u_{inc}$ is varied from $-1$ to $1$. The frequency of the incident light is selected at 294.46 THz, which has been determined in Fig. 1(c). The chiral parameter is chosen as 0.01, 0.02, and 0.03. For $\kappa = 0.01$, the maximum CD can be obtained at $v_{inc}/u_{inc} = -1/26$, which can be well predicted by Eq. (44). Although the ratio of $v_{inc}/u_{inc}$ for maximum CD depends on the chiral parameter, one can also find the optimized excitation condition for different $\kappa$ values by tuning the ratio of $v_{inc}/u_{inc}$. To clearly demonstrate this maximum chiral response, the pure scattering field excited by $E_{inc}$ is varied from $0_1$ to $\pi$, respectively. It can be seen that when $\kappa$ is a complex number, the optimized value $v_{inc}/u_{inc}$ should be searched in the complex plane around the original point to achieve the maximum scattering CD.

In Eq. (9)–(12), the expression of $T_n^a$, $T_n^b$, $T_n^c$, and $T_n^d$ can be expressed as Eqs. (A1)–(A4) [23]:

$$T_n^a = Z_0\psi_n(k_0R_s)\psi_n'(k_1R_s) - Z_1\psi_n(k_1R_s)\psi_n'(k_0R_s)$$

$$T_n^b = Z_0\psi_n(k_1R_s)\psi_n'(k_0R_s) - Z_1\psi_n(k_0R_s)\psi_n'(k_1R_s)$$

$$T_n^c = Z_1k_1\xi_n(k_0R_s)\psi_n'(k_0R_s) - Z_0\psi_n(k_1R_s)\xi_n'(k_0R_s)$$

$$T_n^d = Z_1k_1\xi_n(k_1R_s)\psi_n'(k_0R_s) - Z_0\psi_n(k_0R_s)\xi_n'(k_0R_s)$$

where $R_s$ is the radius of the sphere. $\psi_n(\alpha)$ and $\xi_n(\alpha)$ are the Riccati-Bessel functions.

In Eqs. (26)–(29), the expressions of $S_n^a$, $S_n^b$, $S_n^c$, and $S_n^d$ can be expressed as Eqs. (A5)–(A8):
\[ S_n^\prime = \frac{Z_1 \xi_n^{(1)}(kR_s) \xi_n'(kR_s) - Z_0 \xi_n(kR_s) \xi_n'(kR_s)}{Z_0 \xi_n(kR_s) \psi_n(kR_s) - Z_1 \psi_n(kR_s) \xi_n(kR_s)}, \quad (A7) \]
\[ S_n^\prime = \frac{Z_1 \xi_n^{(1)}(kR_s) \xi_n'(kR_s) - Z_0 \xi_n(kR_s) \xi_n'(kR_s)}{Z_0 \psi_n(kR_s) \xi_n'(kR_s) - Z_1 \psi_n(kR_s) \xi_n'(kR_s)}. \quad (A8) \]

**APPENDIX B: MIRROR OPERATION ON VSH**

The vector spherical harmonics (VSHs) are expressed as [23]

\[ R \hat{M} M_{m,n} = y_m j_n(\rho) \left\{ \sin^m(\theta) \hat{\theta} - \cos^m(\theta) \hat{\phi} \right\} e^{i m \phi}, \quad (B1) \]
\[ R \hat{G} M_{m,n} = -y_m n(n+1) \frac{j_n(kR)}{kR} P_n^m(\cos \theta) e^{i m \phi} \]
\[ + y_m n(n+1) \frac{j_n(kR)}{kR} \left[ \tau_n^m(\theta) \hat{\theta} + i m \pi_n^m(\theta) \hat{\phi} \right] e^{i m \phi}, \quad (B2) \]
\[ M_{m,n} = y_m n(n+1) \frac{h_n^{(1)}(kR)}{kR} P_n^m(\cos \theta) e^{i m \phi} \]
\[ + y_m n(n+1) \frac{h_n^{(1)}(kR)}{kR} \left[ \pi_n^m(\theta) \hat{\phi} \right] e^{i m \phi}. \quad (B4) \]

In Eqs. (B1)–(B4), \( y_m \) is a prefactor for the multipole order of \((m, n)\),

\[ y_m = \sqrt{\frac{(2n+1)(n-m)!}{4\pi n(n+1)(n+m)!}}. \quad (B5) \]

Here, \( j_n \) and \( h_n^{(1)} \) are the spherical Bessel and the first kind spherical Hankel functions. \( P_n^m \) is the associated Legendre function. The functions of \( \pi_n^m \) and \( \tau_n^m \) are defined as

\[ \pi_n^m(\theta) = P_n^m(\cos \theta), \]
\[ \tau_n^m(\theta) = \frac{d P_n^m(\cos \theta)}{d \theta}. \quad (B7) \]

For mirror operation in spherical coordinate, the following transformations are performed simultaneously, i.e., \( r \rightarrow r, \theta \rightarrow \theta, \phi \rightarrow -\phi \). Therefore, \( \hat{M} \hat{R} M_{m,n} = y_m j_n(\rho) i \sin^m(\theta) \hat{\theta} + m \pi_n^m(\theta) \hat{\phi} e^{-i m \phi} \), where \( \hat{M} \) is the mirror operator. The relation between the associated Legendre functions with \((m, n)\) and \((-m, n)\) can be expressed as

\[ P_n^{-m}(\cos \theta) = (-1)^m \frac{(n+m)!}{(n-m)!} P_n^m(\cos \theta). \quad (B8) \]

By using the relations in Eqs. (B1)–(B8), it can be proved that

\[ \hat{M} \hat{R} M_{m,n} = (-1)^{m+1} R \hat{G} M_{m,n}, \quad (B9) \]
\[ \hat{M} \hat{R} N_{m,n} = (-1)^m R \hat{G} N_{m,n}, \quad (B10) \]
\[ \hat{N} M_{m,n} = (-1)^{m+1} M_{m,n}, \quad (B11) \]
\[ \hat{N} N_{m,n} = (-1)^m N_{m,n}. \quad (B12) \]

Equations (B9) and (B10) can be employed to obtain the incident field with the inverted chirality. Therefore, for the incident field of \( E^{(+)} = u_{\text{inc}} R \hat{G} M_{1,1} + v_{\text{inc}} R \hat{G} N_{1,1} \), its mirror field should be \( E^{(-)} = u_{\text{inc}} R \hat{G} M_{-1,1} - v_{\text{inc}} R \hat{G} N_{-1,1} \).

**APPENDIX C: SCATTERING CD OF NANOSPHERE UNDER CPL**

To verify the multiscattering model in this work, the rigorous calculation based on Lorenz-Mie theory is carried out under the illumination of CPL. The \( T \) matrix for the chiral sphere in free space (with the refractive index of \( n_0 \)) can be expressed as [34,35]

\[ T = \begin{bmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{bmatrix}^{-1} \begin{bmatrix} T_{11} \quad T_{12} \\ T_{21} \quad T_{22} \end{bmatrix}. \quad (C1) \]

The elements in Eq. (C1) are given below:

\[ T_{11}^{(1)} = i Z_0 \epsilon_1 \xi_n(kR_s) \psi_n(kR_s) - i \sqrt{\epsilon_1 k_1} \psi_n(kR_s) \xi_n(kR_s), \quad (C2) \]
\[ T_{12}^{(1)} = i Z_0 \epsilon_1 \psi_n(kR_s) \xi_n(kR_s) - i \sqrt{\epsilon_1 k_1} \xi_n(kR_s) \psi_n(kR_s), \quad (C3) \]
\[ T_{21}^{(1)} = -Z_0 \sqrt{\epsilon_1 k_1} \psi_n(kR_s) \xi_n(kR_s) + \mu_1 \xi_n(kR_s) \xi_n(kR_s), \quad (C4) \]
\[ T_{22}^{(1)} = Z_0 \sqrt{\epsilon_1 k_1} \psi_n(kR_s) \xi_n(kR_s) - \mu_1 \psi_n(kR_s) \xi_n(kR_s), \quad (C5) \]
\[ T_{11}^{(2)} = -i Z_0 \epsilon_1 \psi_n(kR_s) \psi_n(kR_s) + i \sqrt{\epsilon_1 k_1} \psi_n(kR_s) \psi_n(kR_s), \quad (C6) \]
\[ T_{12}^{(2)} = -i Z_0 \epsilon_1 \psi_n(kR_s) \psi_n(kR_s) - i \sqrt{\epsilon_1 k_1} \psi_n(kR_s) \psi_n(kR_s), \quad (C7) \]
\[ T_{21}^{(2)} = Z_0 \sqrt{\epsilon_1 k_1} \epsilon_n(kR_s) \psi_n(kR_s) - \mu_1 \epsilon_n(kR_s) \psi_n(kR_s), \quad (C8) \]
\[ T_{22}^{(2)} = -Z_0 \sqrt{\epsilon_1 k_1} \epsilon_n(kR_s) \psi_n(kR_s) + \mu_1 \epsilon_n(kR_s) \psi_n(kR_s). \quad (C9) \]

The two wave vectors inside the sphere are \( k_{LR,R} = \omega \sqrt{\epsilon_s k_s} \pm i \omega k_s \). The radius of the sphere is \( R \). The scattering coefficients can be calculated by \( [a \ b]^T = T [u \ v]^T \). Then the scattering power can be calculated by the scattering coefficients of \( a_{mn} \) and \( b_{mn} \).

In our model, the radius of the sphere is \( R = 320 \text{ nm} \). For \( \epsilon_s = 9, \mu_s = 1, \) and \( k_s = 0.01, \) the scattering CD under the incidence of CPL is calculated as shown in Fig. 6. Within the optical frequency range 176.5–333.3 THz, the value of the scattering CD is in the range \([-0.114, 0.054]\). The analytical method can be used to verify the multiscattering model in this work. For comparison, the scattering CD spectrum calculated by the multiscattering model is shown by the red dots in
Fig. 6. The scattering CD spectrum for chiral sphere under the illumination of CPL. The black curve and the red dots represent the results calculated by the Mie theory and multiscattering model, respectively.

Fig. 7, which proves that the multiscattering model is accurate to describe the chiral response of the Mie sphere with weak chirality.

APPENDIX D: MAXIMUM CD AT 241.46 THz

To explain the mechanism of maximum scattering CD at \( f = 241.46 \text{ THz} \), intensities of the three moments for the sphere and the electric and magnetic scattering coefficients (i.e., \( a_1 \) and \( b_1 \)) are calculated in Fig. 7. The incident field is \( \mathbf{E}^{(1)} = u_{\text{inc}} R_\mathbf{M} \mathbf{e}_{\pm 1,1} + v_{\text{inc}} R_\mathbf{N} \mathbf{e}_{\pm 1,1} \) with \( u_{\text{inc}}/v_{\text{inc}} = 1/55 \). Figures 7(a)–7(c) show the calculation results when the chiral sphere is illuminated by \( \mathbf{E}^{(-)} \). At the frequency of 241.46 THz, both scattering coefficients \( a_1 \) in Fig. 7(a) and \( b_1 \) in Fig. 7(b) are zero, indicating that the sphere is at the completely nonradiating state. Specifically, \( a_1 \) can be well predicted by the amplitude of MD in Fig. 7(a), while \( b_1 \) is mainly contributed by ED and TD (see the amplitudes of \( b_1 \), ED, and TD in Fig. 7(b)). At \( f = 241.46 \text{ THz} \), the amplitudes of ED and TD are equal, but their phases are opposite [see the vertical dashed line through Figs. 7(a)–7(c)], which indicates the excitation of anapole state in the sphere. With the illumination of \( \mathbf{E}^{(+)} \), the amplitudes of ED and TD are almost unchanged as shown in Figs. 7(e) and 7(f). It means the anapole state is stable during the inversion of optical chirality, which is different from the case in Fig. 3. However, by switching the incident field from \( \mathbf{E}^{(-)} \) to \( \mathbf{E}^{(+)} \), the dip of MD amplitude at \( f = 241.46 \text{ THz} \) disappears as shown in Fig. 7(d). Therefore, the maximum CD condition can also be enabled due to the excitation of anapole states.

To further understand the mechanism of the dependence of the magnetic scattering coefficient \( a_1 \) to the chirality of the incident field, Eqs. (32) and (33) should be considered.

to nontraditional chiroptical phenomena, Light Sci. Appl. 9, 139 (2020).


