

Optimal conditions for detecting optical dichroism at the nanoscale by electron energy-loss spectroscopy

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The emergence of optical circular dichroism in chiral nanoscale and molecular systems provides not only a way for analyzing the sample chirality itself but also additional degrees of freedom in manipulating light. Such manipulation can be reached even at the nanoscale level; however, probing and understanding the properties of optical fields well below the diffraction limit requires an adequate technique. Electron energy-loss spectroscopy (EELS) with orbital angular momentum (OAM)-based electron state sorting has been suggested as a suitable candidate, but to date, no conclusive experiments have been evidenced. We, therefore, theoretically explore the emergence of dichroism in EELS for a canonical single-twist helix nanostructure and present a detailed analysis of the optimal parameters to obtain a robust signal. Our work offers insights into the interpretation of the OAM-resolved EELS signal and its sensitivity to various experimental parameters, which can inspire and guide future experimental efforts.

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I. INTRODUCTION

Chirality, the lack of mirror symmetry with respect to any plane that can be considered, is present in many naturally occurring nanoscale systems, such as in amino acids or protein molecules, and it plays an important role in various biological and chemical processes [1]. The chirality can be revealed from direct structural measurements, but it also manifests itself in vibrational or electronic properties, which results in a different response of a chiral molecule to left- versus right-handed circularly polarized (CP) light—circular dichroism [2,3]. The difference in the absorption or emission of CP light by different enantiomers is further utilized in the design of artificially fabricated chiral nanostructures [4–7], which find applications, e.g., in light manipulation [8–13] or in the so-called enhanced spectroscopies relying on the enhancement of the dichroic signal of chiral molecules placed in the tailored optical near field of a chiral nanostructure [14,15].

Although light represents a relatively nondestructive and effective way to probe even sensitive chiral molecules [16], light-based techniques are diffraction limited. Probing the dichroic response from a small number of molecules [17] or even spatial mapping of the dichroic response at subwavelength spatial resolution, which could be particularly useful for understanding the artificially designed chiral near fields

[18,19], thus remains challenging. As an alternative to light, an electron beam represents a suitable probe that can provide spatial resolution down to the single-atom level and simultaneously measure the low-energy vibrational and electronic response of matter [20–23].

It has been theoretically predicted that information on optical and vibrational chirality can be retrieved in electron energy-loss spectroscopy (EELS) experiments that allow for the detection of electrons based on the orbital angular momentum (OAM) transferred in their interaction with the sample [24–29]. Such OAM-based sorting can now be achieved with electron sorters that rely on electrostatic fields [30–32]. One can also consider the possibility of a suitably designed initial electron state that could further enhance the capability of detecting the desired dichroic signal, e.g., a vortex electron beam (VEB) [33,34], which can be generated using approaches based on diffraction [35–37], sculpted thin films [38], or the interaction with static and dynamic electromagnetic fields [39–45].

However, despite the promising theoretical predictions, the experimental detection of nanoscale dichroism due to low-energy excitations in EELS has been inconclusive so far [46], which is even more surprising given the increasing capabilities in generating and detecting states with OAM [32,42]. In this article, we therefore develop a theoretical formalism based on a decomposition of the sample potential in a suitable basis, providing insights into the role of experimental parameters on the resulting dichroic signal. We apply our theoretical findings on a prototypical chiral nanostructure—a plasmonic nanohelix—and demonstrate that dichroism in EELS can exhibit both sign flips and differ over orders of magnitude based on the probing geometry and electron microscope settings.

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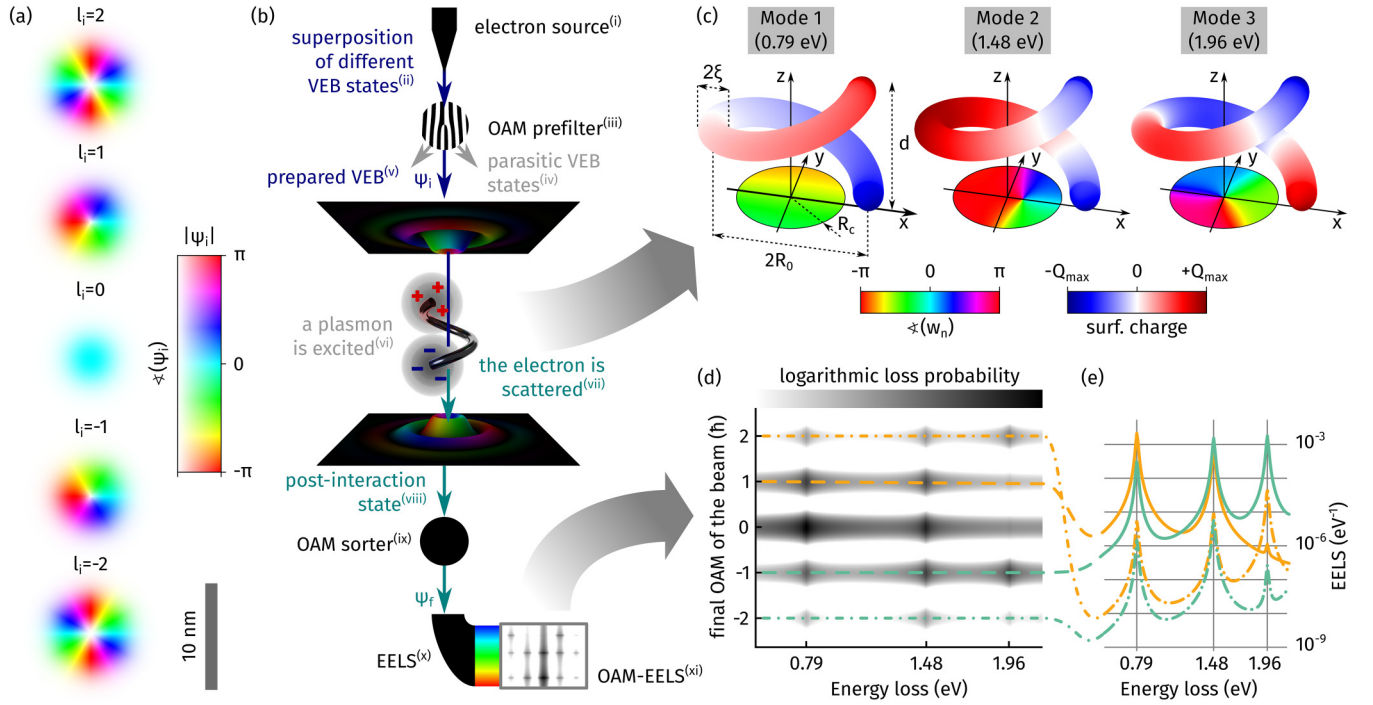


FIG. 1. Overview of the proposed experiment: (a) Phase and absolute values of considered Laguerre-Gauss beams for different OAMs. (b) A scheme of the setup. Electrons from the source (i) in the superposition of different VEB states (ii) pass through an OAM prefilter (iii). A VEB with a well-defined OAM is prepared (v) and the states with different-than-chosen OAM are sorted out (iv). A VEB passing near the sample can excite a plasmon (vi) and scatter (vii). After the interaction, the beam is in a statistically mixed state (viii). An OAM sorter (ix) is used to spatially separate the mixed states. The electrons are further dispersed in energy by the EELS spectrometer (x), forming a 2D OAM-resolved EELS image (xi). (c) Pseudocharge density and phase of the projected eigenpotentials w_n corresponding to the first three modes of a silver helix with thickness $2\xi = 20$ nm, pitch $d = 100$ nm, and radius $R_0 = 50$ nm. (d) Calculated OAM-resolved EELS image for the sample from (c) illuminated by a nonvortex Gaussian beam (energy 30 keV, $\ell_i = 0$, $s_0 = 10$ nm, $\alpha_f = 5$ mrad). The image is smeared in the (vertical) OAM axis by a Gaussian to illustrate the finite resolution of the OAM sorter. (e) A cut through for direct comparison between spectra obtained for final OAMs $\pm\hbar$ and $\pm 2\hbar$, respectively.

II. RESULTS AND DISCUSSION

A. Theoretical framework for OAM-resolved EELS

In the following, we consider a hypothetical experimental setup sketched in Fig. 1(b). We assume that with a suitable method (e.g., using diffraction-based phase plates), we obtain a well-defined initial beam characterized by $\psi_i(R, \phi) = \exp(iq_z z) \exp(i\ell_i \phi) f_{\ell_i}(R)$, where $\hbar q_z$ is the component of the electron momentum along the beam axis (with the reduced Planck constant $\hbar$), $f_{\ell_i}(R)$ is a function dependent on the transverse distance from the origin $R = \sqrt{x^2 + y^2}$, and ϕ is the azimuthal angle. The electrons can have either zero or nonzero topological charge ℓ_i , which quantizes the OAM $L_z = \hbar \ell_i$. See Fig. 1(a), where several transverse VEB profiles with different ℓ_i s are presented. The incident electrons then interact with the sample, and some of them exchange energy or even OAM through the excited near field. The energy loss and OAM transfer can be analyzed afterward using an OAM analyzer and an EELS spectrometer. For instance, the combination of an OAM sorter and an EELS spectrometer [47] could directly yield energy dispersion and OAM dispersion (due to the finite resolution of the OAM sorter) along the two perpendicular axes of the EELS camera as illustrated in Fig. 1(d).

The electron energy-loss probability corresponding to a final state with a specific final OAM $\hbar \ell_f$ can be, in the nonretarded (when the interaction with the sample is described electrostatically) and nonrecoil (when the electron's trajectory is assumed to be unperturbed) approximation, expressed as [25]

$$\Gamma_{\ell_i}^{\ell_f}(\omega) = \frac{e^2}{2\pi^2 \hbar v^2} \sum_{n=1}^{\infty} g_n(\omega) \int_0^{Q_{c,f}} Q_f dQ_f \left| \int R dR \times \int_0^{2\pi} d\phi f_{\ell_i}(R) e^{i\Delta\ell\phi} J_{\ell_f}(Q_f R) w_n(R, \phi, \omega_n) \right|^2, \quad (1)$$

where e is the elementary charge, v is the relativistic electron velocity, $\Delta\ell = \ell_f - \ell_i$ is the topological charge transfer, and we considered that a detector centered at $R = 0$ collects electrons with transverse wave vectors Q_f up to a cutoff $Q_{c,f} = \sin(\alpha_f) \sqrt{2m_e \hbar \epsilon_i (1 + \hbar \epsilon_i / (2m_e c^2))} / \hbar$ corresponding to the collection angle α_f . m_e is the electron mass, $\hbar \epsilon_i$ is the initial electron energy, and c is the speed of light in vacuum. $J_{\ell_f}(x)$ are the Bessel functions of the first kind and order ℓ_f . Properties of the sample excitations are embedded in the functions g_n and w_n , which introduce spectral and spatial dependence,

respectively, of the nanostructure's optical eigenmodes indexed by n (see detailed derivations in Sec. C1, and discussion about the validity of nonretarded approximation in Sec. C3 in the Supplemental Material (SM) [48]). These functions are related to the screened potential $W(\mathbf{r}, \mathbf{r}', \omega)$ as

$$\int dz \int dz' \text{Im}\{-W(\mathbf{r}, \mathbf{r}', \omega)\} e^{-i\omega(z-z')/v} \\ \approx \sum_{n=1}^{\infty} g_n(\omega) w_n(\mathbf{R}, \omega_n) w_n^*(\mathbf{R}', \omega_n),$$

where we considered well-defined, spectrally not-overlapping modes resonating at frequencies ω_n .

We can immediately notice from Eq. (1) that to ensure high spectral magnitudes necessary to overcome noise in EELS experiments, one has to optimize the spatial overlap of the projected sample potential in terms of w_n with the initial and final transverse wave function profiles, which has also been shown in experiments with other types of phase-shaped electron beams [49]. As the optical near fields are typically extended over tens of nanometers, it is therefore desirable to work with not-so-well-focused beams (previously considered in Refs. [50,51]).

We now apply Eq. (1) for the transition from the initial nonvortex state (OAM $\hbar\ell_i = 0$) to the selected final state with OAM $\hbar\ell_f$ in the interaction with a silver single-twist nanohelix [see Fig. 1(c) for the geometry and selected parameters]. The nanohelix is a suitable candidate for demonstrating our formalism because it features clear dichroism when probed by circularly polarized light [52,53]. Moreover, nanohelices can be relatively routinely fabricated in a variety of dimensions using different types of deposition techniques [54–57]. Eigenmodes and the corresponding eigenpotentials of the nanohelix plotted in Fig. 1(c) are calculated in MNPBEM [58], where we also verified validity of the nonretarded approximation and spectral separation of the modes with silver as the helix's material (for details and the optical response of silver, see Sec. C in the SM [48]). We consider a Gaussian transverse profile of the initial wave function with $f_0(R) = \sqrt{2}/(\pi s_0^2) \exp(-R^2/s_0^2)$, where we set the beam waist $s_0 = 10$ nm to minimize interaction of the beam with

the helix's bulk. The calculation is performed for the perfectly aligned axis of the electron beam and nanohelix, for an electron microscope operated at the acceleration voltage of 30 kV (electron velocity $v = 0.328c$) with a collection angle $\alpha_f = 5$ mrad, which are feasible parameters for EELS in a scanning transmission electron microscope. Figure 1(d) summarizes the spectra calculated for different ℓ_f s that could be distinguished using the OAM sorter. In analogy to dichroism in light optics, the dichroism in EELS emerges for a nonzero difference between spectra acquired for $+\Delta\ell$ and $-\Delta\ell$ (as discussed in detail in Sec. II C). The selected spectra obtained for $\Delta\ell = +1, +2$ and $\Delta\ell = -1, -2$ plotted separately in Fig. 1(e) exhibit clear differences and therefore demonstrate the emergence of dichroism in EELS. For instance, the first peak at 0.79 eV is more intense for $\Delta\ell = +1$ and $+2$. Importantly, we can notice that the magnitude of the EELS signal rapidly decreases with increasing $\Delta\ell$ [note the logarithmic scale in Figs. 1(d) and 1(e)], which already indicates that high-OAM transitions might not be favorable when probing optical dichroism.

B. Analytical model for a single-twist nanohelix

To further understand the results presented in Fig. 1 and potentially improve the efficiency and robustness of the dichroic signal, it is beneficial to develop an analytical model. We employ the decomposition of the spatial eigenpotential dependence w_n in the Fourier-Bessel basis, which is particularly suitable due to the cylindrical symmetry of the system:

$$w_n(R, \phi, \omega_n) = \sum_{m=-\infty}^{\infty} i^m e^{im\phi} \sum_{j=1}^{\infty} J_m\left(\alpha_{j,m} \frac{R}{a}\right) A_{j,m}^n, \quad (2)$$

where m are integers, a is a finite radius of an area, where we evaluate the projected potential, and $A_{j,m}^n$ are expansion coefficients expressed as

$$A_{j,m}^n = \frac{(-i)^m}{\pi a^2 [J_{m+1}(\alpha_{j,m})]^2} \int_0^a R dR J_m\left(\frac{\alpha_{j,m} R}{a}\right) \\ \times \int_0^{2\pi} d\phi e^{-im\phi} w_n(R, \phi, \omega_n), \quad (3)$$

where $\alpha_{j,m}$ denotes j th zero of the Bessel function of order m . After substituting Eq. (2) into Eq. (1), we find

$$\Gamma_{\ell_i}^{\ell_f}(\omega) = \frac{2e^2}{\hbar v^2} \sum_{n=1}^{\infty} g_n(\omega) \int_0^{Q_c} Q_f dQ_f \left| \int_0^{R_c} R dR f_{\ell_i}(R) J_{\ell_f}(Q_f R) \sum_{j=1}^{\infty} J_{\Delta\ell}\left(\alpha_{j,\Delta\ell} \frac{R}{a}\right) A_{j,-\Delta\ell}^n \right|^2, \quad (4)$$

which immediately shows that to observe the emergence of a dichroic signal, the coefficients $A_{j,\Delta\ell}$ have to be different for $+\Delta\ell$ and $-\Delta\ell$. For numerical calculations, we used $a = 5R_0$, and cutoff radius $R_c = 0.7R_0$.

In the case of a nanohelix, we can proceed with the evaluation of the coefficients (see Sec. B1 of the SM [48]) if we approximate the distribution of charges corresponding to the resonant eigenmodes with a cosine dependence $\cos(n\phi/2)$. Such dependence has been previously derived for linear plasmonic antennas [59], and experiments suggest that it is preserved even for bent nanorods [60]. This approximate charge distribution applied for the helical antenna indeed yields an excellent agreement between analytically and numerically calculated w_n when evaluated further away from the nanoparticle boundaries (see Fig. S6

in the SM [48]). The loss probability then becomes

$$\Gamma_{\ell_i}^{\ell_f}(\omega) = \frac{8e^2}{\hbar} \sum_{n=1}^{\infty} \frac{\mathcal{N}_n^2 g_n(\omega)}{v^2} \int_0^{Q_c} Q_f dQ_f \left| \sum_{j=1}^{\infty} \frac{J_{\Delta\ell}(\alpha_{j,\Delta\ell} \frac{R_0}{a}) + \alpha_{j,\Delta\ell} I_{\Delta\ell}(\frac{\omega_n R_0}{v}) J_{\Delta\ell+1}(\alpha_{j,\Delta\ell}) K_{\Delta\ell}(\frac{\omega_n a}{v})}{(\alpha_{j,\Delta\ell})^2 + (\frac{\omega_n a}{v})^2} \right. \\ \left. \times \int_0^{R_c} R dR f_{\ell_i}(R) J_{\ell_f}(Q_f R) \frac{J_{\Delta\ell}(\alpha_{j,\Delta\ell} \frac{R}{a})}{[J_{\Delta\ell+1}(\alpha_{j,\Delta\ell})]^2} \right|^2 \underbrace{\left| \frac{(1 + (-1)^{1+n} e^{-it_n})(t_n/(2\pi) - \Delta\ell)}{(t_n/(2\pi) - \Delta\ell)^2 - (n/2)^2} \right|^2}_{\mathcal{F}_{\Delta\ell}(t_n)}, \quad (5)$$

where $\mathcal{N}_n = \frac{\xi}{\sigma_n} \sqrt{R_0^2 + (\frac{d}{2\pi})^2}$ with σ_n being the normalization constant obtained from MNPBEM simulation (see Sec. C2 of the SM [48] for details), and 2ξ , d , and R_0 are thickness, pitch, and radius of the nanohelix, respectively, as marked in Fig. 1(c). I_m and K_m are modified Bessel functions of the first and second kind, respectively, and order m . More importantly, Eq. (5) [as well as Eq. (1)] depends primarily on $\Delta\ell$ rather than the exact values of ℓ_i and ℓ_f . The sole dependence on the initial OAM is through the incident wave function's radial profile f_{ℓ_i} . Interestingly, from the azimuthal integral in Eq. (3) emerges function $\mathcal{F}_{\Delta\ell}(t_n)$ dependent on a parameter $t_n = \omega_n d/v$. The function contains information about the electron beam (v), the helix geometry (d), the optical mode (n), and the change of the OAM ($\Delta\ell$). It is also straightforward to see that there exist t_n s for which the function $\mathcal{F}_{\Delta\ell}$ differs from $\mathcal{F}_{-\Delta\ell}$. This is the only term that is changed when flipping the sign of $\Delta\ell$ in Eq. (5). See profound discussion and derivation in Sec. B1 and Sec. B2 of the SM [48]. This condition, indicating the emergence of dichroism, can be fulfilled for all modes.

C. Relative and absolute dichroism

To evaluate the dichroic signal in EELS, we first focus on the relative dichroism, defined as the relative difference between the EEL probability for a transition $\ell_i \rightarrow \ell_f$ and its symmetric counterpart $-\ell_i \rightarrow -\ell_f$, resulting in a compact expression when taking advantage of Eq. (5) and evaluating at energies of individual modes

$$\mathcal{RD}_{\ell_i}^{\ell_f}(\omega_n) = \frac{\mathcal{F}_{\Delta\ell}(t_n) - \mathcal{F}_{-\Delta\ell}(t_n)}{\mathcal{F}_{\Delta\ell}(t_n) + \mathcal{F}_{-\Delta\ell}(t_n)}. \quad (6)$$

The relative dichroism is particularly useful for comparing dichroic spectra among different initial parameters despite potentially large differences in spectral magnitudes. Furthermore, here it is solely governed by $\Delta\ell$ and t_n within $\mathcal{F}_{\Delta\ell}(t_n)$. By substituting for $\mathcal{F}_{\Delta\ell}(t_n)$ in Eq. (6) and considering only positive $\Delta\ell$ (negative $\Delta\ell$ yields opposite results) and small positive t_n (corresponding to a small right-handed structures probed at tens of keV), we can analytically determine the values of t_n that yield maximal (± 1) or zero relative dichroism. When studying helical particles with fixed geometry (constant d), maximal or minimal relative dichroism can be achieved by changing the acceleration voltage of the probe (i.e., changing the velocity v). It is therefore crucial to know in which region of the parametric space we probe.

The resultant t_n dependence of the relative dichroism analysed in Fig. 2 is governed by the comparison of n and $2\Delta\ell$. For $n < 2\Delta\ell$ [e.g., Figs. 2(a1) and 2(b1)–2(b3)], the relative dichroism starts increasing to positive values, reaching its

first maximum at $t_n = \pi(2\Delta\ell - n)$ (see Sec. B2 in the SM [48] for detailed algebra). The change in sign of the relative dichroism shown in Fig. 2 is a significant feature of the system and emphasizes the importance of the theoretical analysis and interpretation. For $n > 2\Delta\ell$ [Fig. 2(a3)] the behavior is even more complex. The relative dichroism starts decreasing to negative values, reaching its first minimum at $t_n = \pi(n - 2\Delta\ell)$. Importantly, after this minimum, the relative dichroism does not return to zero but continues to a second minimum at $t_n = 2\pi\Delta\ell$, forming a “plateau.” For $n = 3$ and $\Delta\ell = 1$, this plateau results in the relative dichroism remaining very close to -1 over a wide range of t_n parameters. When $n = 2\Delta\ell$ [Fig. 2(a2)], a special case with only two extrema (one minimum and one maximum) can occur.

Although the relative dichroism is often used as a measure of the dichroic signal even in light-optics experiments, it has a significant drawback: it can yield maxima even for a very

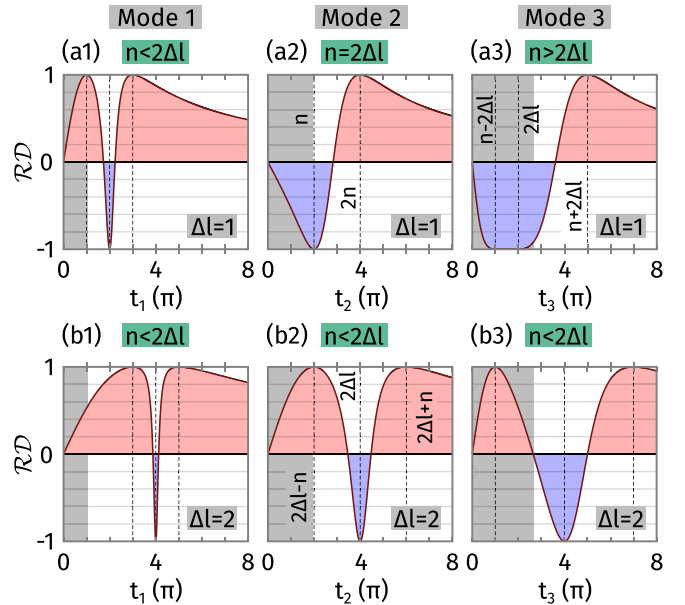


FIG. 2. Relative dichroism dependence on the parameter $t_n(\omega_n, d, v)$ for the first three modes (each column corresponds to one mode). The upper row (a1)–(a3) is calculated for $\Delta\ell = 1$ while the lower row (b1)–(b3) for $\Delta\ell = 2$. Notice the similarities in panels (a1) and (b1)–(b3), and a qualitatively different behavior in (a2) and (a3). The values of t_n for maximal and minimal $\mathcal{RD}$ are displayed in (a2), (a3), and (b2). Shaded regions depict relevant t_n intervals for the first three modes studied in Figs. 3(b1)–3(c3). Vertical axes span from -1 to $+1$ in all graphs.

small spectral response corresponding to very few scattering events. Particularly in EELS, which is fundamentally limited by the Poisson (shot) noise and where also other noise sources can arise [61], one can obtain poor signal-to-noise ratios (SNRs). It might therefore be desirable to rely on the absolute dichroism defined as $\mathcal{AD}_{\ell_i}^{\ell_f}(\omega) = \Gamma_{\ell_i}^{\ell_f}(\omega) - \Gamma_{-\ell_i}^{-\ell_f}(\omega)$, which reflects absolute values of the scattering probabilities. By substituting Eq. (5) and evaluating the $\mathcal{AD}$ at the energies of individual modes, we obtain

$$\begin{aligned} \mathcal{AD}_{\ell_i}^{\ell_f}(\omega_n) &= \frac{8e^2\mathcal{N}_n^2[\mathcal{F}_{\Delta\ell}(t_n) - \mathcal{F}_{-\Delta\ell}(t_n)]}{\hbar v^2} g_n(\omega_n) \\ &\times \int_0^{Q_c} Q_f dQ_f \left| \sum_{j=1}^{\infty} \mathcal{Z}_{j,\Delta\ell} \int_0^{R_c} R dR f_{\ell_i}(R) \right. \\ &\times \left. J_{\ell_f}(Q_f R) J_{\Delta\ell} \left(\alpha_{j,\Delta\ell} \frac{R}{a} \right) \right|^2, \end{aligned} \quad (7)$$

which is again strongly governed by the functions $\mathcal{F}_{\Delta\ell}(t_n)$, but also scaled by the integral featuring spatial dependence of the wave functions. For brevity, we used a function $\mathcal{Z}_{j,\Delta\ell}$ defined by Eq. (S17) in the SM [48]. It is clear from the definitions of dichroism in Eqs. (6) and (7) that it can appear only if the electron transfers OAM to the sample, i.e., $\Delta\ell \neq 0$.

We compare the relative and absolute dichroism for a silver thin helix in Fig. 3(a). We assume the simplest experimental configuration, featuring an initially Gaussian electron beam with $\ell_i = 0$. Selecting the lowest possible OAMs ($\ell_f = \pm 1$) we obtain the EEL spectra for the positive (Γ_0^1) and negative (Γ_0^{-1}) difference of OAM, and corresponding absolute $\mathcal{AD}_0^1$ and relative $\mathcal{RD}_0^1$ dichroism. From Eq. (6) and for $\Delta\ell = 1$ we see that the intensity of the relative dichroism for the first mode is always positive. On the other hand, it is negative for the second and third modes, which can be explained by the probed t_n regions visualized as the shaded areas in Figs. 2(a1)–2(a3). The third mode exhibits $\mathcal{RD}$ very close to the value -1 for the investigated region of helix pitches and electron energies (see Fig. S3(c3) for lower energies in the SM [48]). By evaluating the parameter t_3 , we can indeed find that we probe a large part of a “plateau” area in Fig. 2(a3), where the analytical model yields $\mathcal{RD} \approx -1$.

Next, we focus on the dependence of $\mathcal{AD}$ on some of the relevant experimental parameters, which can help determine whether the signal is measurable with one’s experimental equipment. Figures 3(b1)–3(b3) shows the intensity maps of $\mathcal{AD}_0^1$ as a function of the acceleration voltage and helix pitch for the first three modes [we note that the blue rectangles represent the peak values from Fig. 3(a)]. Qualitatively following the predictions obtained from the analysis of $\mathcal{RD}$, the optimal parameters are different for each mode. Figures 3(c1)–3(c3) show the intensity maps of $\mathcal{AD}_0^2$ for the same parameters as in the previous subfigure. Note the rapid decrease of intensity for all modes compared to $\mathcal{AD}_0^1$ in Figs. 3(b1)–3(b3).

Similar results can be seen in Figs. 4(a1)–4(a3), although the incident beam is now Laguerre-Gaussian with $\ell_i = 1$. Because the OAM difference is the same as in Figs. 3(b1)–3(b3), the absolute dichroism is only scaled by a different value of the overlap integral in Eq. (7). In this case, $\mathcal{AD}_1^2$ is approximately twice as large compared to $\mathcal{AD}_0^1$. This be-

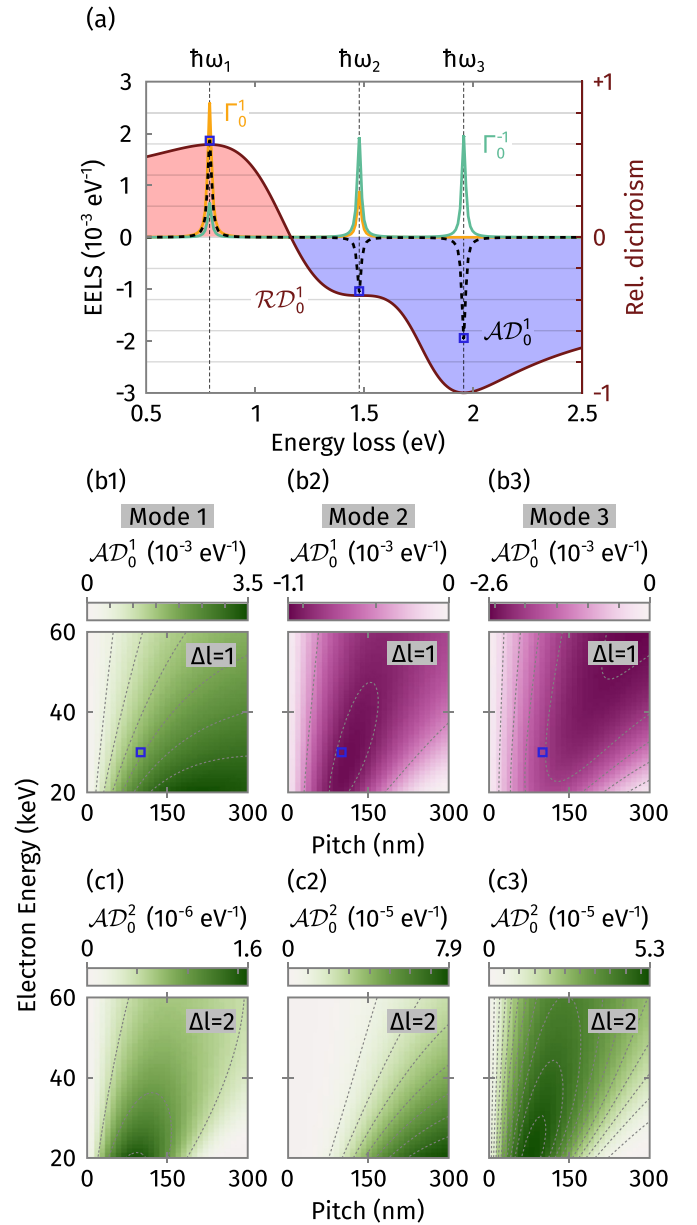


FIG. 3. Dichroism for an infinitesimally thin silver single-twist helix [$R_0 = 50 \text{ nm}$; a thickness of $\xi = 10 \text{ nm}$ was used to determine the normalization constant σ_n from MNPBEM simulation; pitch is $d = 100 \text{ nm}$ in (a) and varies in (b1)–(d3)] perfectly aligned with an electron beam [waist $s_0 = 10 \text{ nm}$; energy is 30 keV in (a) 20 – 60 keV in (b), (c)]. We consider the collection of all electrons, thus $Q_c \rightarrow \infty$. (a) EEL spectra for $\ell_i = 0$ and positive (Γ_0^1 ; yellow) and negative (Γ_0^{-1} ; green) OAM exchange with corresponding absolute dichroism ($\mathcal{AD}_0^1$; dashed black) and relative dichroism ($\mathcal{RD}_0^1$; maroon). (b) Absolute dichroism for the first three modes as a function of the helix pitch and electron energy for the OAM transition $0 \rightarrow \pm 1$. The contour step is $5 \times 10^{-4} \text{ eV}^{-1}$, and the blue rectangles indicate the values of the absolute dichroism in (a). (c) Same as (b) for $\ell_i = 1$ and the transition in OAM $1 \rightarrow \pm 2$. Ticks in colorbars mark the contour levels.

havior can be intuitively explained by the donutlike region with a large beam intensity emerging for the $\ell_i = 1$ beam, which is closer to the helix boundaries and thus excites the nanoparticle more efficiently. Further optimization of the

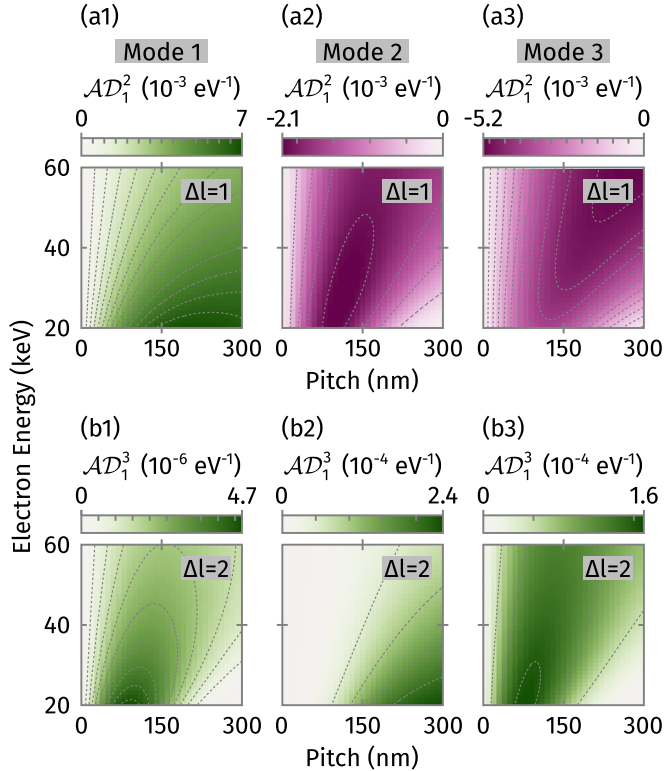


FIG. 4. Dichroism for an infinitesimally thin silver single-twist helix ($R_0 = 50$ nm; a thickness of $\xi = 10$ nm was used to determine the normalization constant σ_n from MNPBEM simulation; pitch varies from 0 to 300 nm) perfectly aligned with an electron beam (waist $s_0 = 10$ nm; energy varies from 20 to 60 keV in all graphs). We consider the collection of all electrons, thus $Q_c \rightarrow \infty$. (a) Absolute dichroism for the first three modes as a function of the helix pitch and electron energy for the OAM transition $1 \rightarrow \pm 2$. The contour step is $5 \times 10^{-4} \text{ eV}^{-1}$. (b) Same as (a) but for the OAM transition $1 \rightarrow \pm 3$. The contour steps are 5×10^{-7} , 5×10^{-5} , and $5 \times 10^{-5} \text{ eV}^{-1}$, respectively. Ticks in colorbars mark the contour levels.

resulting $\mathcal{AD}$ could be performed by varying beam convergence (s_0) and collection conditions (see Fig. S2 in the SM [48], where we show the dependence on collection angle, OAM transition, and beam width). We also note that even though the $\mathcal{RD} \approx -1$ for $n = 3$ and almost all pitches and electron energies under consideration [see Fig. S3(c3)], the $\mathcal{AD}$ for low energies (10–20 keV) and large pitches (300 nm) is close to zero—the dichroic signal would be very difficult to obtain experimentally [see Fig. S3(a3)].

In Figs. 4(b1)–4(b3) we explore the case of $\Delta\ell = 2$ by evaluating $\mathcal{AD}_1^3$, which reaches positive values for all three modes [we note that $\mathcal{AD}_0^2$ behaves similarly in Figs. 3(c1)–3(c3) except for a different scaling]. This can be easily predicted by looking at the shaded region in Figs. 2(b1)–2(b3). Again, one can see even bigger increase in intensity for VEB in $\mathcal{AD}_1^3$ in Figs. 4(b1)–4(b3) compared to ordinary Gaussian beams in $\mathcal{AD}_0^2$ in Figs. 3(c1)–3(c3) due to the high beam intensity closer to the nanoparticle boundaries. However, the transition $1 \rightarrow 3$ provides approximately an order of magnitude lower value of $\mathcal{AD}$ compared to the cases with $\Delta\ell = 1$.

We also find that the sign of the dichroism within the parametric space of interest can help to determine the handedness of the helix. For $\Delta\ell = 2$, we can assign the nanohelix as right handed (resp. left handed), the absolute (and relative) dichroism is positive (resp. negative) for the lowest three modes [see Figs. 3(c) and 4(b)]. Alternatively, dichroism for $\Delta\ell = 1$ is positive/negative/negative for the right-handed helix and the modes 1/2/3 as shown in Figs. 3(b) and 4(a). Such analysis, however, cannot be easily generalized to any chiral nanoparticle as it is possible only thanks to the knowledge of the function $\mathcal{F}_{\Delta\ell}$ and its dependence on the parameter t_n . We also note that even for the nanohelix it becomes more complex when we extend the parametric space (see Fig. S3 [48]).

So far, we have considered an idealized scenario in which an electron beam passes exactly through the axis of the helix. However, one can suspect that under realistic experimental conditions, the beam can be misaligned—shifted or tilted as sketched in Figs. 5(a) and 5(c), respectively. We again utilize the analytical expression for the helix’s eigenpotentials, but due to the disturbed symmetry, we evaluate Eq. (1) numerically. Importantly, when probing the first three modes, the absolute dichroism $\mathcal{AD}_0^1$ only shows quantitative changes for both beam shift [Figs. 5(b1)–5(b3)] and tilt [Figs. 5(d1)–5(d3)]. These findings hold not just for the lowest transition with $\Delta\ell = 1$ and an incident Gaussian beam ($\ell_i = 0$) (with $s_0 = 10$ nm, $\alpha_f = 5$ mrad, and energy 30 keV), but also for other initial and final electron states (see Figs. S4 and S5) and $\mathcal{RD}$ again proving that $\Delta\ell$ is the critical parameter determining behavior of the dichroic signal. However, in certain cases [see, e.g., Fig. 5(b2)], the dichroic signal can change quantitatively, which could be crucial when fighting with SNR in the experiment, and, more importantly, a nonzero dichroic signal can emerge even when probing an achiral nanostructure. If the system of a beam and a probed object, e.g., a “C-shaped” nanostructure [pitch $d = 0$ nm; see a schematic in Fig. 5(e)] under a tilt is chiral, an “extrinsic” dichroism in EELS emerges as demonstrated in Figs. 5(f1)–5(f3).

III. CONCLUSIONS

We present a theoretical framework that allows for quantitative evaluation of the optical dichroic signal in electron energy-loss spectroscopy. Using an example of a plasmonic nanohelix, we demonstrate that both the sign and magnitude of the dichroic signal are governed not only by geometrical parameters of the sample and properties of its optical modes, but also by the exchanged OAM ($\Delta\ell$) and electron energy. We find analytical conditions to maximize the relative dichroism, analogous to phase-matching conditions for maximizing EELS with conventional electron beams [62–64], and demonstrate the importance of theoretical analysis by showing the complex dependence of the dichroic signal over the parameter space.

We also focus on the influence of geometrical misalignments that may affect experiments. We show that, within a reasonable level of precision, the dichroic signal should be robust with respect to shifts and tilts between the sample and beam axes. However, it is still important to check the dependence of the signal on the tilt, as a nonzero, yet small

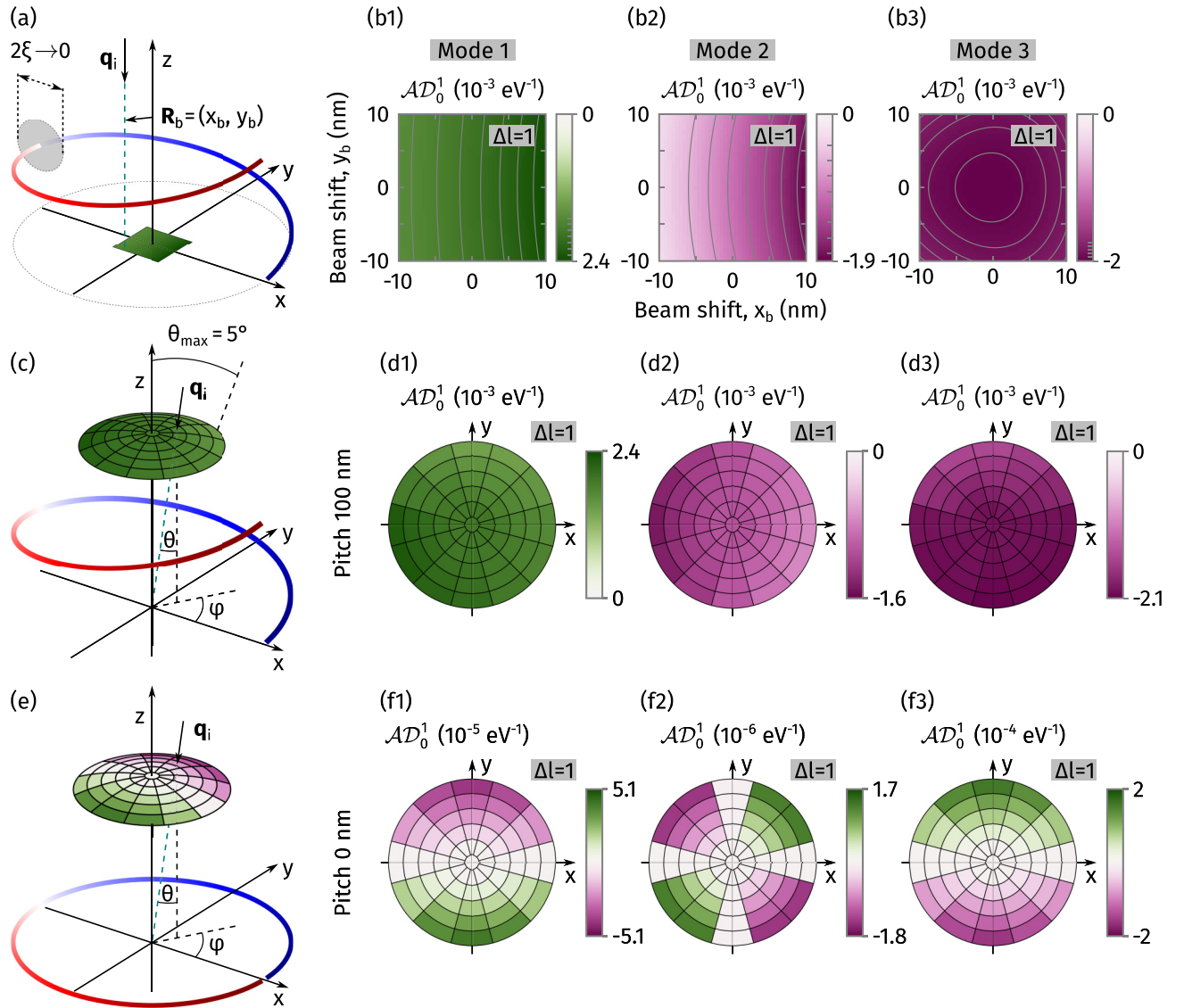


FIG. 5. Absolute dichroism $\mathcal{AD}_0^1$ for an infinitesimally thin silver single-twist helix [$R_0 = 50$ nm, $\xi = 10$ nm, $d = 100$ nm in (b), (d) and $d = 0$ nm in (f)] probed by a 30 keV Gaussian electron beam with $s_0 = 10$ nm, and $\alpha_f = 5$ mrad when the electron beam is scanned or tilted. (a) Scheme of the “scanning” simulation, where the beam’s trajectory is parallel to the helix axis, impinging at position $\mathbf{R}_b = (x_b, y_b)$. $\mathcal{AD}$ is plotted in the 20×20 nm computation area in (b1)–(b3) for the first three modes. The contour steps are $\{1; 2; 0.5\} \times 10^{-4} \text{ eV}^{-1}$, respectively. (c) Scheme of the “tilting” simulation for the beam tilt represented in the spherical coordinate system by angles $(\theta; \varphi)$. The electron beam passes through the origin of the coordinate system. Each “pixel” in the circular “lid” represents a unique impinging direction of the beam. The results are computed for polar angles θ ranging from 0° to 5° with a step 1° . The azimuthal dependence is divided into 12 parts. (d1)–(d3) $\mathcal{AD}_0^1$ for the first three modes when tilting the beam. (e) Scheme of the “tilting” simulation for a “C-shaped” structure ($d = 0$ nm). (f1)–(f3) Same as (d1)–(d3) but for a “C-shaped” structure ($d = 0$ nm) exhibiting extrinsic dichroism.

and volatile, extrinsic dichroic signal can emerge for a tilted achiral sample. We further suggest that it is preferable to focus on low OAM transfers and small ℓ_i , ensuring the best spatial overlap with the projected sample potential, which typically varies on the nanoscale level when dealing with optical excitations. In such a scenario, one can achieve higher magnitudes of absolute dichroism, effectively overcoming issues with low signal-to-noise ratios. We note that the presented magnitudes could be further enhanced by optimizing the incident electron beam profile for an optimal overlap with the projected sample potential. However, we proved that dichroism can also be

studied by nonvortex beams, but the OAM postselection is crucial in both cases.

Our work contributes to the establishment of new EELS techniques that utilize state postselection and symmetry matching to actively control selection rules and acquire more detailed information on sample excitations [26,29,49]. Particularly, we set the ground for near-future measurements of optical dichroism in setups that allow postselection of electron states based on OAM. Such experiments could help to understand the OAM transfer and behavior of chiral optical fields at the nanoscale.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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