

Chiral patterning of rough surfaces with vortex laser beams: from structured polarization to twisted forces

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The ability to create surface structures with precisely controlled chirality remains a major challenge in laser-matter interaction experiments. In this work, we theoretically study the interaction of vortex laser beams, characterized by spiral polarization patterns and twisted wavefronts, with rough metallic surfaces in order to create surface patterns with chirality. Using numerical simulations based on the finite-difference time-domain method, we investigate how spin and orbital angular momenta influence the inhomogeneous energy absorption at the surface and generate twisted optical forces that can drive topographic reorganization. We show how different structured light fields can create intricate patterns with chiral features on a material surface. We emphasize the crucial role of polarization and spatial inhomogeneity of the light field in the generation of asymmetric torque forces that directly affect the surface dynamics. Our electromagnetic simulations show how vortex beams can be used to create chiral surface structures, expanding our knowledge of laser-generated periodic surface structures and opening up new possibilities for chiral surface engineering.

I. INTRODUCTION

The interaction of intense laser radiation with surfaces of solids leads to the emergence of laser-induced periodic surface structures (LIPSS), which significantly modify both the topographical and functional properties of the irradiated materials [1]. These periodic undulations, first observed in the mid-1960s [2], have seen a surge in research interest following the advent of ultra-short laser pulses [3]. LIPSS can be viewed as surface ripples with varying depth and periodicity, the formation of which is influenced by the spatiotemporal coherence and polarization of the laser pulse [4, 5]. The process of LIPSS formation and their properties are affected by a number of parameters, including pulse duration [6], laser fluence [7], polarization direction [5], wavelength [8, 9], and the number of applied pulses [10]. LIPSS can be generated on the surface of a wide variety of materials, including metals [11–13], semiconductors [3, 8, 14, 15], glasses [16, 17] and polymers [18–20], demonstrating the versatility and broad applicability of LIPSS in various areas of materials science. Coherent irradiation of multiple points on a rough surface initiates a complex interaction between various physical mechanisms, ranging from interference between scattered waves [21] to near-field enhancement effects [22], optical resonances [22, 23] such as surface plasmons [24, 25], and activation of hydrodynamic instabilities [26]. As a result of these processes, local temperature gradients arise, which, through a thermo-mechanical response, lead to the formation of a modulated surface relief with certain axes of symmetry. Traditionally, LIPSS are divided into two classes: low-spatial-frequency LIPSS (LSFL), which often occur

near the ablation threshold, and high-spatial-frequency LIPSS (HSFL), which are driven by thermo-convective effects [27]. Typically, these surface structures have only one axis of symmetry, which is determined by the polarization of the laser pulse. However, recent experiments have demonstrated that using multiple time-delayed laser pulses, it is possible to produce surface structures with two or even three axes of symmetry, creating complex patterns such as cross-hatching or hexagonal lattices [28]. In addition to the pursuit of maximum miniaturization, one of the central challenges of laser surface processing has become the creation of new surface structures with unusual geometry [25, 29].

The compelling question now is: can we go beyond conventional symmetry and create surface structures with fully asymmetric patterns, independent of laser polarization direction? Typically, chiral patterns lacking mirror symmetry naturally possess these geometric properties and thus compare favorably with currently created LIPSS. It is conceivable that such chiral patterns could be produced using laser pulses that have their own intrinsic chirality. In turn, the intrinsic chirality of laser pulses is closely related to the ability of light to have angular momentum. Light can carry angular momentum in two forms: spin angular momentum (SAM) and orbital angular momentum (OAM) [30, 31]. SAM is associated with circular polarization of light and manifests itself in the form of two discrete states: left-hand and right-hand circular polarization, where the polarization handedness determines the sign of the angular momentum. In contrast, OAM has a continuous range of values determined by the so-called topological charge, which can be any positive or negative integer. The laser pulses carrying the OAM have a doughnut-like shape and a spiral wavefront [30]. Although SAM has received much more attention in LIPSS research, studying the effects of OAM may reveal new patterns and lead to new functional surfaces. For further study of chiral interactions of light and matter, it is ex-

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tremely important to take into account not only spatially changing polarization, but also spatially inhomogeneous phase of structured light beams [32]. The ability to create chiral surface structures will not only revolutionize the fundamental understanding of LIPSS formation, but will also open the way to innovative applications in such diverse fields as chiral molecular sensing [33], enantiomer separation [34], disease diagnosis and treatment [35, 36], and chiral light manipulation [37].

When irradiating an isotropic surface, the geometry of LIPSS is primarily determined by the properties of the incident laser pulses: beam shape, laser polarization, and wavefront geometry. By tuning the intensity distribution across the beam profile and using non-Gaussian beam shapes such as flattop, Bessel or Laguerre-Gauss, we gain precise control over the spatial placement of LIPSS. In turn, the orientation of LIPSS within the laser spot can be controlled by changing the spatial distribution of the laser pulse polarization. In particular, by using vector beams with spatially varying polarization, we can create LIPSS with intricate patterns such as azimuthal, radial or spiral [38–40]. The creation of chiral surface patterns using structured optical beams via direct surface irradiation has been reported [41]. Recent experiments with vector beams have demonstrated polarization-directed formation of helical nanostructures [42]. This phenomenon utilizes self-aligning of near-field enhancement, which causes the growth of surface structures oriented along the polarization vector [5]. By surface processing with laser pulses having radial and azimuthal polarization distributions, it is possible to create large areas of complex biomimetic structures [43, 44]. Finally, specially designed wavefronts, such as those with a twisted shape and associated with OAM, can induce the formation of spiral surface formations. For example, during photopolymerization or ablation, laser pulses with such wavefronts twist the temporarily molten material, creating a chiral surface morphology as it solidifies [45–47].

Currently, two scenarios are proposed to explain the physical mechanism underlying the OAM-induced helical surface morphologies: hydrodynamic and electrodynamic [47]. The first scenario is based on helical gradients of temperature and surface tension arising from interference between the incident OAM beam and its replica [46]. This interference creates a chiral temperature pattern and surface tension profiles at the surface, resulting in helical thermocapillary motion of the molten material. This mechanism allows the surface to be structured by directly imprinting the gradients of absorbed energy during the melting process. The second scenario involves direct transfer of angular momentum to the rotational motion of the molten material under the influence of radiation forces [45, 48]. This idea suggests the presence of an electrodynamic torque responsible for the chiral motions of the material. According to this hypothesis, the helical morphology of the surface should reflect the direction of the wavefront rotation. Despite the existence of the two scenarios, one important question remains: can

these scenarios be used to explain the interaction of short femtosecond laser pulses with surfaces that transition to a liquid phase (the only phase in which molten material can move) only a few picoseconds after laser irradiation? It is also difficult to create a sufficiently thick liquid layer (more than a few wavelengths) because the short duration of the laser pulse means that the heating cannot be maintained for long enough. Finally, in the absence of standing waves, it is reasonable to ask about the significance of the effect of averaging the action of the rotating wave front and the corresponding optical forces over several optical periods.

To explore the possibilities of creating new surface patterns, in this paper we study the influence of OAM and SAM on LIPSS formation. Using finite-difference time-domain (FDTD) method, we simulate the interaction of structured light fields with rough surfaces. We consider laser pulses with different OAMs, polarization states, and wavefront geometries to predict and control the resulting LIPSS patterns. In our approach we use a statistical description of surface roughness to analyze the distribution of absorbed laser energy and to demonstrate how different structured light fields affect the properties of LIPSS. Our results show that structured light fields with helical polarization, spiral intensity distribution, or twisting optical forces can create surface structures with curved geometries and spatial arrangements. These effects, likely more significant than energy gradients unaffected by OAM, significantly expand the control available in laser manufacturing.

II. THE NUMERICAL MODEL

To study the interaction of structured light pulses with rough surfaces, we numerically solve the following system of Maxwell equations using the FDTD method [49]:

$$\vec{\nabla} \times \vec{E} = -\frac{\partial \vec{B}}{\partial t}, \quad \vec{\nabla} \times \vec{H} = \frac{\partial \vec{D}}{\partial t}, \quad (1)$$

where $\vec{E}(\vec{r}, t)$ and $\vec{H}(\vec{r}, t)$ are the electric and magnetic field vectors, $\vec{r} = \{x, y, z\}$ is the coordinate vector and $\vec{B} = \mu_0 \vec{H}$, with μ_0 being the vacuum permeability. The medium response can be expressed through the displacement field $\vec{D}(\vec{r}, t)$ written in the frequency domain as $\tilde{D}(\vec{r}, \omega) = \varepsilon_0 \varepsilon(\omega) \tilde{E}(\vec{r}, \omega)$, where \sim denotes the temporal spectrum, ε_0 is the vacuum permittivity and $\varepsilon(\omega)$ is the frequency-dependent permittivity of the medium.

As the source of radiation, we consider a laser pulse launched from an xy plane in the $-z$ direction (like in an experiment, from top to bottom). We can express the electric field vector \vec{E} of such laser pulse through its $E_x(\vec{r}, t)$ and $E_y(\vec{r}, t)$ components as

$$E_x = E_{x0} \cos \theta - E_{y0} \sin \theta, \quad (2a)$$

$$E_y = E_{x0} \sin \theta + E_{y0} \cos \theta, \quad (2b)$$

where

$$E_{x0} = \frac{1}{\sqrt{1+\epsilon^2}} A(x, y, t) \cos(\omega_0 t + \phi(x, y)), \quad (3a)$$

$$E_{y0} = \frac{\epsilon}{\sqrt{1+\epsilon^2}} A(x, y, t) \sin(\omega_0 t + \phi(x, y)). \quad (3b)$$

Here $A(x, y, t)$ is the spatio-temporal amplitude, $\phi(x, y)$ is the phase, and ω_0 is the central frequency, such that $\lambda_0 = 2\pi c_0 / \omega_0$ is the central wavelength with c_0 being the speed of light in vacuum. The parameter ϵ defines the ellipticity of the polarization and equals to the ratio of the semi-axes of the polarization ellipse. In particular, $\epsilon = 0$ and $\epsilon = 1$ correspond to, respectively, linear and circular polarizations. Different signs of ϵ define the left-hand and right-hand polarizations. The factor $1/\sqrt{1+\epsilon^2}$ in Eq. (3) guarantees that $E_x^2 + E_y^2$ remains constant, independently of ϵ , i.e. that laser pulses of different polarization have the same energy. Although Eqs. (3) allow us to obtain polarizations of arbitrary ellipticity, the orientation of the polarization ellipse is fixed: the major semi-axis is always coincides with the x direction. Therefore, in order to introduce the polarization of arbitrary orientation, in Eq. (2) we multiply the field components E_{x0} and E_{y0} by the rotation matrix where the rotation angle θ is measured relative to the positive direction of the x axis. With the spatially varying polarization ellipticity ϵ and angle θ we can define inhomogeneous polarization states of arbitrary complexity.

In our simulations we assume that the laser pulse can be represented by a plane wave with the temporal envelope defined as a one period of \sin^2 function:

$$A(x, y, t) = A_0 \sin^2 \left(\frac{\pi}{2} \frac{t}{\tau_0} \right), \quad (4)$$

where A_0 is the peak amplitude of the electric field and τ_0 is the full width at half maximum pulse duration. Since the beginning and the end of the \sin^2 pulse in time are well-defined, we can save a significant amount of computation time by avoiding modeling slowly rising pulse front and tail, like, for example, in the case of the Gaussian envelope. To take into account that any phase $\phi(x, y)$ other than flat one changes the amplitude distribution in time, we also apply the temporal transformation where in Eq. (4) we replace the time t by $t + \phi(x, y)/\omega_0$ (see Appendix A). In our simulations we do not consider any intensity-dependent effects of laser-matter interaction and, therefore, without loss of generality, we take $A_0 = 1$ V/m. Additionally, we assume the pulse duration $\tau_0 = 100$ fs and the central wavelength $\lambda_0 = 1.03$ μm .

Below the radiation plane, we place a semi-infinite stainless steel medium. To model the dispersive response of stainless steel we apply the auxillary differential equation method [49] assuming the Drude permittivity $\varepsilon(\omega) = 1 - \omega_p^2/(\omega^2 + i\omega\gamma)$ with the plasma frequency $\omega_p = 19.2 \times 10^{15}$ 1/s and the damping rate $\gamma = 9.15 \times 10^{15}$ 1/s [50]. With these parameters the complex refractive index $n = n' + in''$ of stainless steel

at $\lambda_0 = 1.03$ μm has the real and imaginary parts equal to $n' = 3.02$ and $n'' = 3.51$ with the corresponding skin depth $l = 1/(2n''\omega_0/c_0) = 23.34$ nm.

Since LIPSS originate from the interference of incoming light and light asymmetrically scattered at surface inhomogeneities [21], as well as from contributions of nonradiative field enhancement on roughness [50], it is essential to account for the rough surface of the stainless steel sample. To enhance realism, we assume that the rough surface has a continuous distribution of heights that can be statistically described. In particular, the surface roughness is represented by the function $R(x, y)$, which defines the random deviations of the surface height relative to a reference plane [51]. To express the statistical properties of the surface roughness we use the correlation function $C(X, Y) = \langle R(x, y)R(x+X, y+Y) \rangle / \sigma^2$, where $\langle \dots \rangle$ denotes the spatial averaging and $\sigma = \sqrt{\langle R^2 \rangle}$ is the root-mean-square (rms) surface height. The correlation function $C(X, Y)$ describes the spatial coherence between surface heights at different points separated by the distance $d = \sqrt{X^2 + Y^2}$. In our simulations we assume the Gaussian correlation function $C(X, Y) = \sigma^2 \exp(-(X^2 + Y^2)/\xi^2)$, where ξ is the correlation length. For details on converting this correlation function into an actual roughness function $R(x, y)$, refer to [52–54]. In our simulations we use the rms height $\sigma = 50$ nm and the correlation length $\xi = 100$ nm, which are approximately ten times smaller than the laser wavelength. These values correspond to a well-polished surface with subwavelength inhomogeneities required for HSFL observation.

The computational grid in our FDTD simulations has the sizes $L_x=L_y=14.6$ μm and $L_z=1.2$ μm in the x , y , and z directions, respectively, with the corresponding step sizes $\Delta x=\Delta y=10$ nm and $\Delta z=5$ nm. To avoid nonphysical reflections, at each end of the grid we place convolutional perfectly matched layers of 0.1 μm thickness. The stainless steel medium is placed 0.6 μm below the radiation plane (as measured relative to the reference plane of the roughness function $R(x, y)$). Figure 1 illustrates the resulting surface roughness $R(x, y)$ on our computational grid.

In our studies of LIPSS formation, our main interest is focused on the distribution of laser energy absorbed at the surface. The surface areas that have absorbed a sufficiently large amount of energy will be extruded in the process of the laser-matter interaction and, thus, act as a seed for the growth of LIPSS [25, 26]. With the electric field $\vec{E}(x, y, z, t)$ obtained by the FDTD simulations we can calculate the energy W delivered by the laser pulse per unit volume as $W(x, y, z) = 2\varepsilon_0 n'' \omega_0 \int_{-\infty}^{\infty} E^2(x, y, z, t) dt$ [52, 55]. Then, in terms of our coordinate system, the distribution of the energy $Q(x, y)$ absorbed by the surface of stainless steel can be calculated as the integral of $W(x, y, z)$ over all z layers of the surface: $Q(x, y) = \int_{-\infty}^{\infty} W(x, y, z) dz$. Finally, the total laser energy Q_{tot} absorbed by the surface can be obtained as $Q_{\text{tot}} = \iint_{-\infty}^{\infty} Q(x, y) dx dy$. The distribution

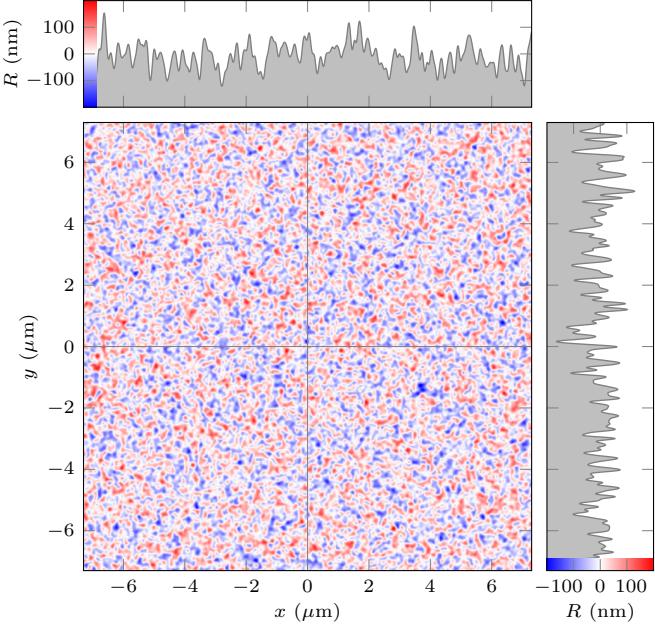


FIG. 1. The surface roughness $R(x, y)$ (the distribution of surface heights relative to the $z = 0$ plane) used in our simulations. The line plots show the corresponding cross-sections at $y = 0$ (top) and $x = 0$ (right).

of absorbed laser energy $Q(x, y)$ allows us to predict the resulting geometry of LIPSS, while the total absorbed energy Q_{tot} allow us to compare the strength of light-surface coupling for different laser pulses.

III. SEARCHING FOR CHIRAL LIPSS

A. Linear and circular polarizations

As a starting point, let us consider a laser pulse that has linear polarization oriented along the x direction ($\epsilon = 0$ and $\theta = 0^\circ$ in Eqs. (2) and (3)). Figure 2(a) shows the distribution of the laser energy $Q(x, y)$ absorbed by the stainless steel sample irradiated by such laser pulse. The surface roughness causes the distribution of $Q(x, y)$ to resemble a chaotic pattern of absorbed energy spots. However, upon closer inspection we find that the regions of high absorption are elongated in the direction of laser polarization. Taking into account that the regions of high losses act as a seed for LIPSS growth, we can expect that the resulting LIPSS will be also oriented in the x direction — parallel to the laser polarization.

To obtain more information about the orientation and size distribution of the absorbed energy spots, we calculate the spatial spectrum of $Q(x, y)$. Figure 2(d) shows the spectrum of $Q(x, y)$ in the spatial-frequency coordinates k_x and k_y normalized by the wave number $k_0 = \omega_0/c_0$. We see that the spatial spectrum of $Q(x, y)$ has a well-recognizable shape with spectral features known as "type-s" and "type-r" [26]. We also see

that, compared to the type-s features, oriented along the k_x direction, the type-r features, oriented along the k_y direction, consist of spectral components with higher frequencies. Considering that smaller shapes in space correspond to higher spectral frequencies, we can conclude that the spots of absorbed energy are indeed, on average, compressed in the y direction and stretched in the x direction. The spatial spectra of $Q(x, y)$ also provide information on the typical size of the absorbed energy spots. We can use this information to estimate the period of the resulting LIPSS and to distinguish LSFL from HSFL. The spectral components of $Q(x, y)$ located at spatial frequencies close to or less than k_0 are responsible for the formation of LSFL since they correspond to large spots with characteristic sizes less than or equal to the laser wavelength λ_0 . In turn, the spectral components of $Q(x, y)$ at spatial frequencies much larger than k_0 correspond to the small-scale sub-wavelength spots responsible for the formation of HSFL. In Fig. 2(d) we see that the highest frequency components of $Q(x, y)$ spectrum lie in the region of $5k_0$ which means that the minimum size of the absorbed energy spots is approximately five times smaller than the laser wavelength λ_0 . Thus, we can predict that the minimum period of the resulting LIPSS will be $\lambda_0/5$.

Next, let us consider the same linearly polarized laser pulse but with the polarization rotated by 45 degrees relative to the x axis ($\theta = 45^\circ$ in Eq. (2)). Figures 2(b) and (e) show the corresponding distribution of absorbed energy $Q(x, y)$ and its spatial spectrum. From the comparison of Figs. 2(d) and (e) we see that the spectrum of $Q(x, y)$ in Fig. 2(e) is rotated by 45 degrees, which means that the orientation of the corresponding absorbed energy spots is also changed. A closer look at the absorbed energy spots in Fig. 2(b) shows that they are indeed elongated along the polarization direction. Thus, our simulations confirm the known fact that the orientation of LIPSS follows the direction of laser polarization.

Finally, let us consider a circularly polarized laser pulse ($\epsilon = +1$ and $\theta = 0^\circ$ in Eqs. (2) and (3)). Figures 2(c) and (f) show that in this case the distribution of absorbed laser energy $Q(x, y)$ does not have any preferred orientation and the corresponding spatial spectrum is symmetrical about the origin. This observation can be explained by the fact that for a circularly polarized laser pulse the electric field vector rotates, passing through all possible orientations. Therefore, even if at each moment in time the laser pulse has a certain direction of polarization, the result of laser-matter interaction becomes averaged over all polarization angles. Thus, we can expect that in the case of a circularly polarized laser pulse, the corresponding LIPSS will have a symmetric shape without a preferred orientation.

We note that for all three polarization cases the total amount of absorbed laser energy Q_{tot} is the same (the difference is less than numerical errors). We also checked that for a circularly polarized laser pulse with the opposite direction of polarization rotation ($\epsilon = -1$) the distri-

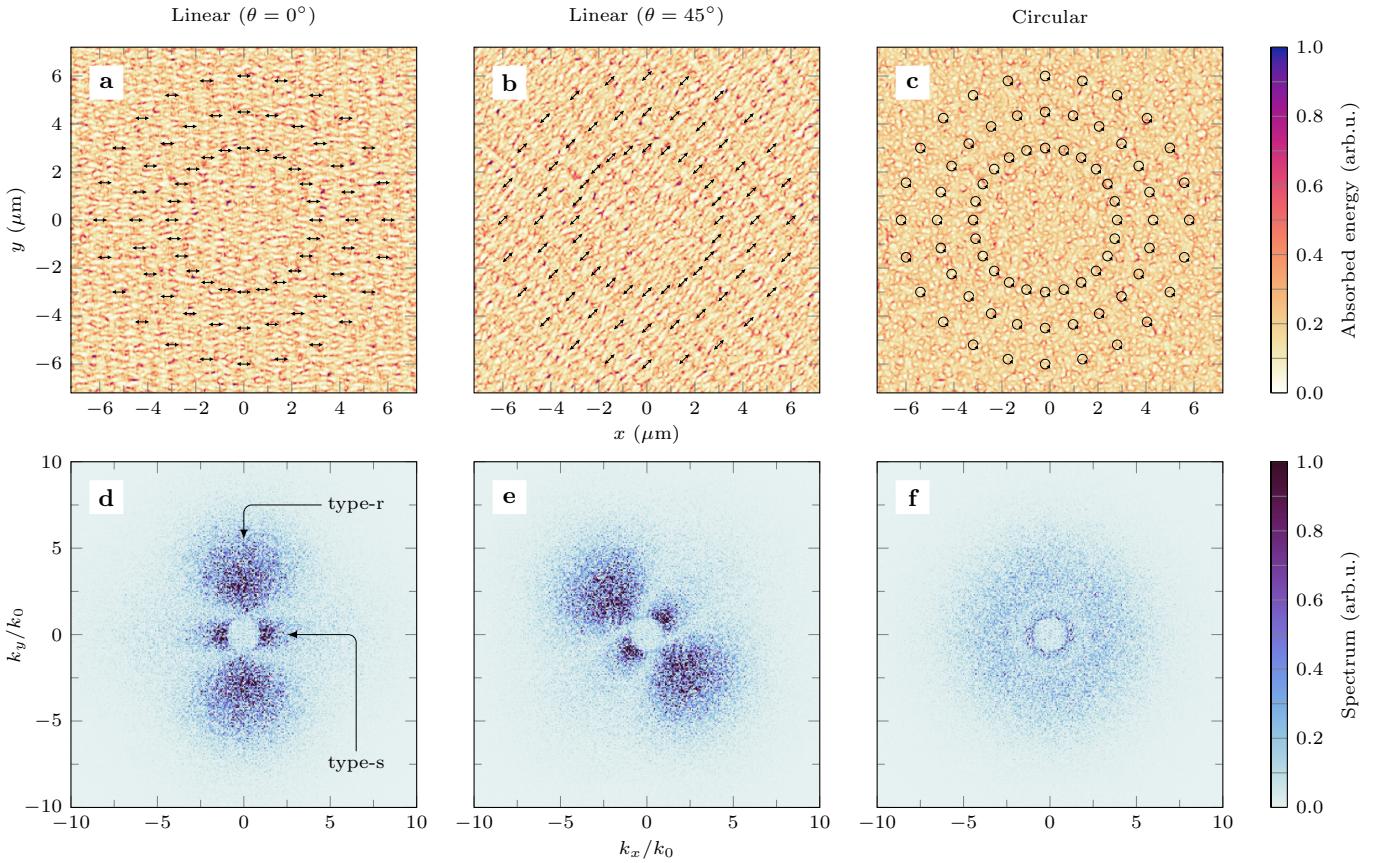


FIG. 2. The distributions of absorbed energy $Q(x, y)$ (a,b,c) and their spectra (d,e,f) for laser pulses with linear polarization rotated by $\theta = 0^\circ$ (a,d) and $\theta = 45^\circ$ (b,e), and for a circularly polarized laser pulse (c,f). The arrows in (a–c) show the direction of the laser polarization. The arrows in (d) mark the characteristic spectral patterns known as "type-r" and "type-s" features.

bution of absorbed energy $Q(x, y)$ changes insignificantly, while the total losses Q_{tot} remain unchanged, indicating the absence of circular dichroism. Thus, we can conclude that the morphology of resulting LIPSS does not depend on the polarization handedness and, consequently, on the sign of the SAM.

B. Inhomogeneous polarization distribution

As we have just seen, for linearly polarized laser pulses the distribution of absorbed laser energy $Q(x, y)$ looks like a set of elongated spots aligned along the polarization direction, which allows us to assert that the laser polarization determines the orientation of the resulting LIPSS. We can exploit this dependence on the polarization direction to obtain LIPSS with complex morphology. For this purpose we can use laser pulses with inhomogeneous polarization distribution, where the local polarization direction will determine the orientation of LIPSS at a given point. As an example, let us consider three laser pulses with the radial, spiral, and azimuthal polarization patterns. In terms of Eqs. (2) and (3) such laser pulses are defined by the linear polarization

with $\epsilon = 0$ and the spatially-dependent polarization angle $\theta(x, y) = \arctan(y/x) + \psi$, where $\psi \in [-\pi/2, \pi/2]$ determines the angle between the polarization direction and the radius vector of a given point with the coordinates x and y . In particular, $\psi = 0$ and $\psi = \pm\pi/2$ correspond to the radial and azimuthal polarization patterns, respectively, while the intermediate values of ψ define the spiral polarization patterns of different vorticity (the sign of ψ allows us to switch between the left-handed and right-handed rotation of the spiral). Experimentally, such laser pulses can be generated, for example, using so-called q-plates [56]. Figure 3 shows the distributions of absorbed laser energy $Q(x, y)$ and their spectra obtained for laser pulses having the radial ($\psi = 0$), spiral ($\psi = \pi/4$), and azimuthal ($\psi = \pi/2$) polarization patterns as a result of their interaction with the rough stainless steel surface. The arrows in Figs. 3(a)–(c) allow us to visualize the distribution of the polarization for each of the patterns. Figures 3(a)–(c) show that, as expected, locally the spots of absorbed laser energy are oriented along a given polarization direction, forming a distribution that repeats the polarization pattern; the central region with zero absorption appears due to the zero on-axis intensity caused by the polarization singularity at this point. Since the regions

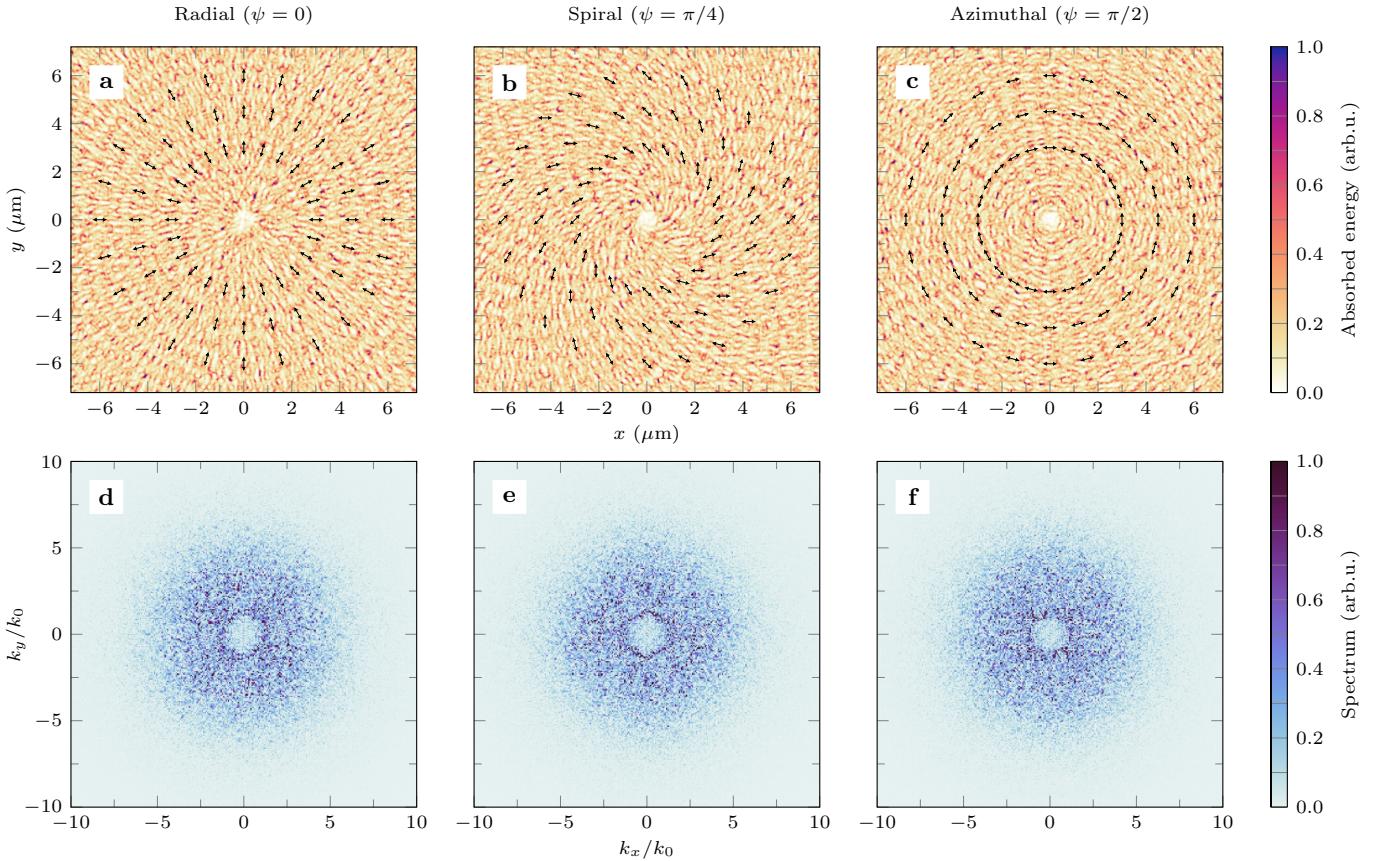


FIG. 3. The distributions of absorbed energy $Q(x, y)$ (a,b,c) and their spectra (d,e,f) for laser pulses with inhomogeneous polarization distribution defined by the polarization rotation angle $\theta(x, y) = \arctan(y/x) + \psi$ with $\psi = 0$ for the radial (a,d), $\psi = \pi/4$ for the spiral (b,e), and $\psi = \pi/2$ for the azimuthal (c,f) polarization patterns. The arrows in (a–c) indicate the direction of the laser polarization in a given point.

of high losses act as seeds for the formation of LIPSS, the resulting LIPSS will be organized in accordance with the polarization pattern. Such intricate LIPSS formations, obtained using complex polarization states, have already been observed in several experiments [38–40]. Thus, the laser pulses with spiral polarization distributions allow us to create LIPSS patterns with controllable vorticity.

Interestingly, Fig. 3(d)–(f) show that the spectra of $Q(x, y)$ for the radial, spiral, and azimuthal polarizations are practically identical and resemble the spectrum obtained for the circularly polarized laser pulse (see Fig. 2(f)). We can explain this observation by the fact that for each of the three polarization patterns there are regions in the beam cross section containing all possible orientations of linear polarization. As a result, the final spectrum can be seen as a set of $Q(x, y)$ spectra for a linearly polarized laser pulse (see Figs. 2(d) and (e)), averaged over all possible angles of polarization.

During the simulation, we did not find any significant difference in the total absorbed energy Q_{tot} between the laser pulses with the radial, spiral, and azimuthal polarization — the maximum difference did not exceed 0.5%. We also verified that the value of Q_{tot} does not change by any noticeable amount for laser pulses having the

spiral polarization with an opposite handedness given by $\theta(x, y) = \arctan(y/x) - \pi/4$. Thus, we can conclude that the total amount of absorbed laser energy is insensitive to the polarization vorticity.

C. Orbital angular momentum

In the previous section we saw that it is possible to create chiral LIPSS formations using inhomogeneous polarization states. Let us now explore the possibility of creating chiral LIPSS patterns using laser pulses carrying OAM. According to the Sipe's theory [21] LSFL arise as a result of interference between incident laser radiation and surface electromagnetic waves generated by scattering on a rough surface. Therefore, we can expect that for laser pulses with OAM, their interference with the light scattered on the surface will form a pattern of interference maxima which will inherit the helical structure of the wavefront and which, being imprinted on the surface in the form of regions of high absorption, will lead to emergence of chiral LIPSS. In order to explore this possibility we consider a linearly polarized laser pulse ($\epsilon = 0$ and $\theta = 0^\circ$ in Eqs. (2) and (3)) with a helical phase

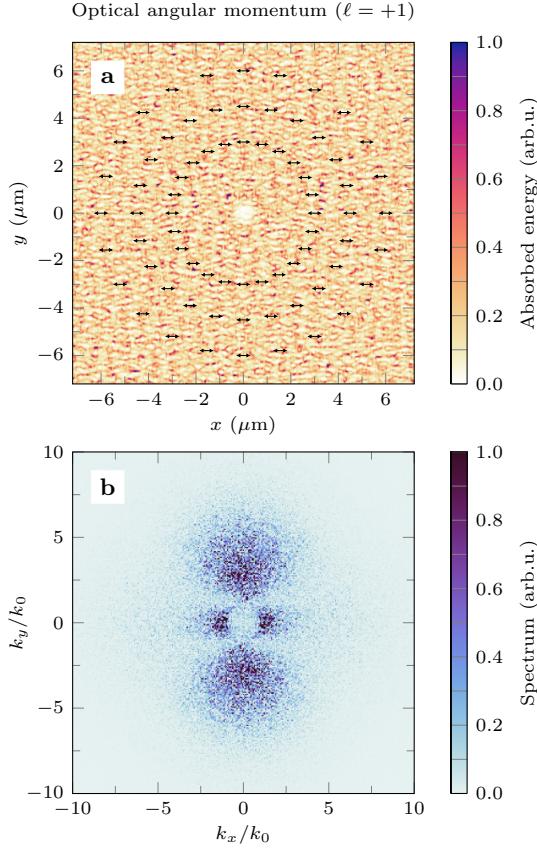


FIG. 4. The distribution of absorbed energy $Q(x, y)$ (a) and its spectrum (b) for the linearly polarized laser pulse carrying OAM with the topological charge $\ell = +1$. The arrows in (a) show the direction of the laser polarization.

$\phi(x, y) = \ell \arctan(y/x)$, where ℓ is an integer number known as the topological charge. The magnitude of ℓ dictates the number of rotations the wavefront undergoes in one period of the laser pulse, while the sign of ℓ indicates the direction of this rotation. Figure 4 shows the distribution of absorbed laser energy $Q(x, y)$ and its spectrum for the laser pulse having the OAM with $\ell = +1$. In Fig. 4(a) we see that the $Q(x, y)$ distribution does not show any traces of vorticity and is very similar to the distribution obtained for a linearly polarized laser pulse with a flat phase (see Fig. 2(a)): the only difference is the region of zero losses in the center, corresponding to the zero on-axis intensity caused by the phase singularity at that point. In turn, a comparison of Fig. 4(b) with Fig. 2(d) shows that the spectra of the laser pulse with OAM and a conventional linearly polarized laser pulse are practically identical. In our studies we also considered laser pulses with higher values (up to 10) and different signs of the topological charge ℓ . However, the shape of the resulting distribution of absorbed laser energy did not show any traces of chirality. Thus, we can conclude that the presence of OAM in the laser pulse does not affect the distribution of absorbed laser energy and, therefore, does not change the geometry of LIPSS.

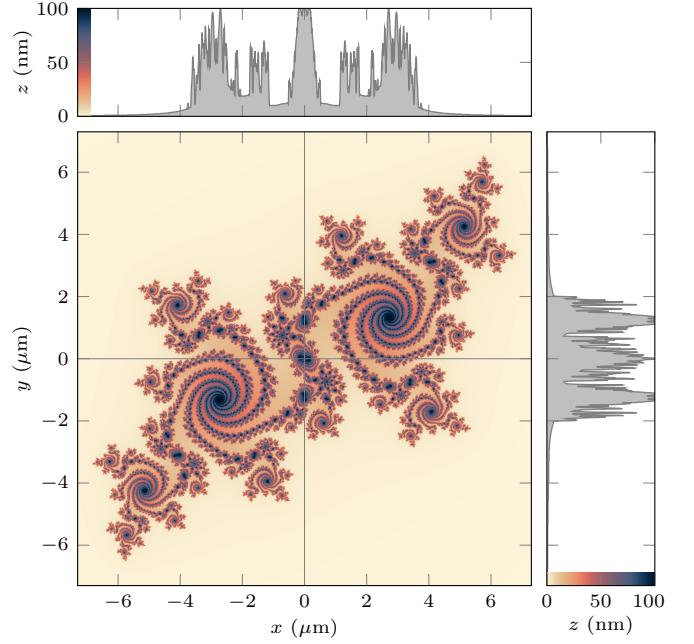


FIG. 5. Geometry of the fractal structure on the surface of stainless steel used to study the response of a surface with its own chirality. The line plots show the cross-sections of the fractal structure at $y = 0$ (top) and $x = 0$ (right).

D. Fractal surface structures

As we saw above, contrary to our expectations, when a laser pulse interacts with a rough surface, the presence of OAM does not change the distribution of the absorbed laser energy. However, how might the surface's own chirality affect the interaction with the rotating wavefront in an hypothetical context of helical dichroism? From experiment we know that at least individual chiral nanostructures are capable of responding differently to the sign of the OAM in the incoming laser pulse [57]. Therefore, we can expect that the distribution of absorbed laser energy on a surface containing some chiral structures will be different for laser pulses with different OAMs. But what kind of chiral structures should we choose to maximize the response to incoming laser pulses? Of course, we could conduct a parametric study by playing with the size, shape, and arrangement of an array of chiral nanoparticles deposited on the surface. However, we decided to simplify the problem by taking a surface with a fractal chiral structure applied to it. As such fractal structure we consider the Julia set $J(f)$ defined from the function $f(z) = z^2 + c$ with $c = -0.5125 + 0.5213i$ and protruding 100 nm above the flat surface. Figure 5 shows the resulting geometry of the fractal stainless steel surface used in our simulations. We see that the fractal nature of the Julia set allows us to obtain a surface structure consisting of chiral elements whose scale starts at a few wavelengths and gradually decreases to sub-wavelength sizes. As a result, in one simulation run we are able to

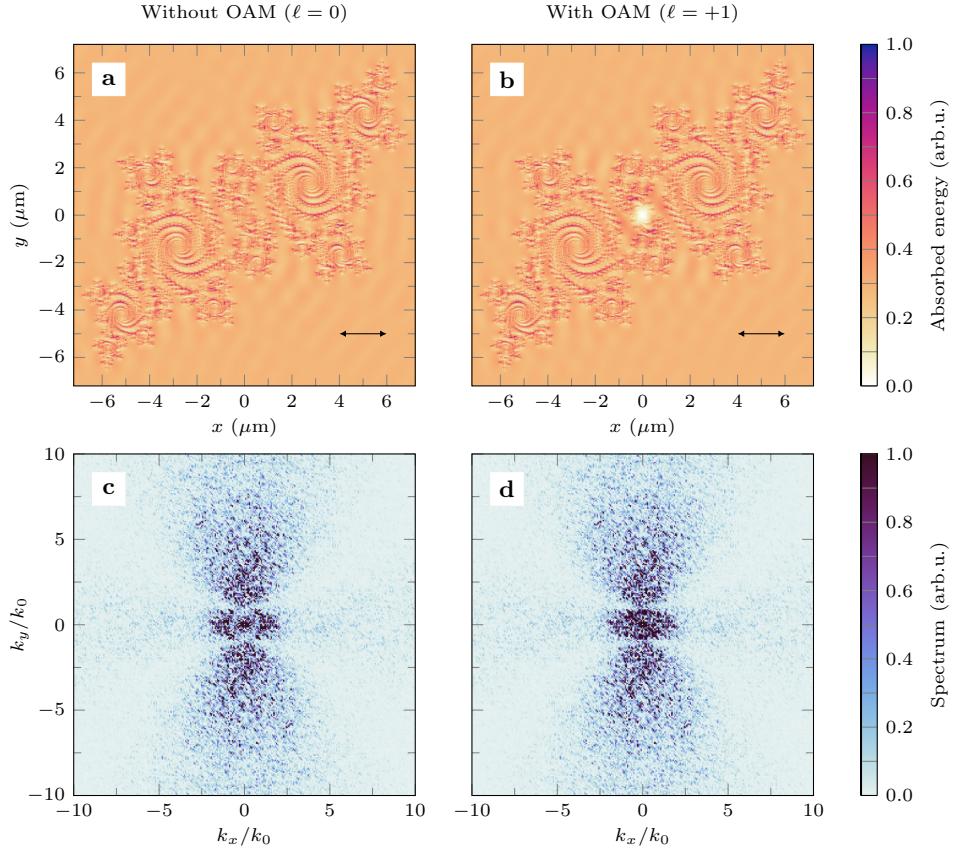


FIG. 6. The distributions of absorbed energy $Q(x, y)$ (a,b) and their spectra (c,d) for linearly polarized laser pulses interacting with the fractal surface structure. (a) Laser pulse without OAM ($\ell = 0$) and (b) with OAM of topological charge $\ell = +1$. The arrows in (a,b) show the direction of the laser polarization.

scan a whole range of chiral structures of different scales, some of which will have to be in resonance with the incident radiation.

Figure 6 shows the distributions of absorbed energy $Q(x, y)$ and their spectra obtained as a result of the interaction with the fractal surface of linearly polarized laser pulses without ($\ell = 0$) and with ($\ell = +1$) OAM. In Figs. 6(a) and (b) we see that, apart of the central region in Fig. 6(b) with zero losses caused by the phase singularity, both distributions of $Q(x, y)$ do not contain any significant differences visible to the naked eye that could distinguish the cases of $\ell = 0$ and $\ell = +1$. In Fig. 6(c) and (d) we see that the spectra of $Q(x, y)$ contain more high-frequency spectral component compared to the case of a linearly polarized laser pulses interacting with the rough surface (see Fig. 2(d)). This is because, compared to a rough surface, the fractal structure consists of much smaller scatterers. The only visible difference between the spectra in Fig. 6(c) and (d) is observed in the region of zero frequencies and is due to the presence of a zero-loss spot in Fig. 6(b). We verified that changing the sign of the topological charge ℓ does not lead to any visible changes. We also tested laser pulses with OAM of higher topological charges (up to $\ell = \pm 10$), but did not find any effect of OAM. Additionally, our simulations show that

the total absorbed energy Q_{tot} is insensitive to the sign of the topological charge ℓ , independently of the amplitude of ℓ . Thus, we can argue that the presence of chiral structures on the surface does not guarantee that the distribution of absorbed energy, and therefore the geometry of resulting LIPSS, will sense the presence of OAM. Furthermore, in the context of positive feedback from repeated laser pulses at the same location, there should be no enhancement of the chiral effect.

E. Spiral intensity distribution

So far, we have not identified a configuration where a laser pulse with OAM produces chiral distributions of absorbed laser energy that could induce the formation of chiral LIPSS. However, instead of seeking a direct OAM effect, we can exploit OAM indirectly to create a spiral intensity distribution. We can expect that such intensity distribution, being imprinted on the surface, will lead to a spiral arrangement of LIPSS. To obtain the spiral intensity pattern we can superimpose a focused OAM laser pulse with a second one having a plane wave front [58, 59]. To recreate such a combination of laser pulses in our simulations, we use the sum of

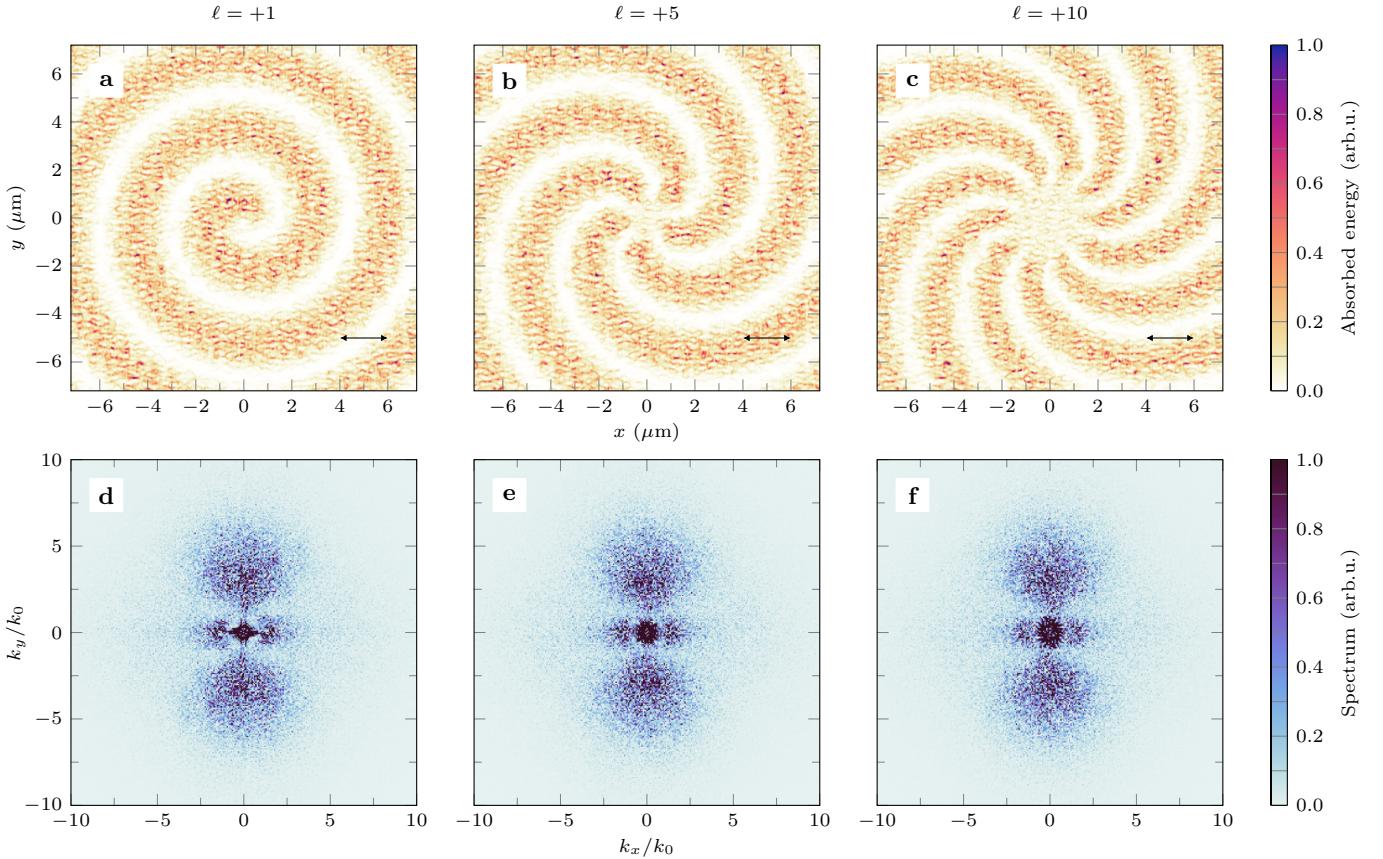


FIG. 7. The distributions of absorbed energy $Q(x, y)$ (a,b,c) and their spectra (d,e,f) for linearly polarized laser pulses with spiral intensity distribution obtained using OAMs with topological charges $\ell = +1$ (a,d), $\ell = +5$ (b,e), and $\ell = +10$ (c,f). The arrows in (a,b,c) show the direction of the laser polarization.

a plane-wave x -polarized laser pulse ($\epsilon = 0$ and $\theta = 0^\circ$ in Eqs. (2) and (3)) and a laser pulse with the phase given by $\phi(x, y) = \ell \arctan(y/x) - k_0 \sqrt{x^2 + y^2} \sin \delta$, where the first term defines the helical wave front associated with the OAM and the second term describes a conical phase intended to simulate tight focusing with a convergence angle of δ . We assume that $\delta = 20^\circ$, which corresponds to the focusing with the numerical aperture $NA = \sin \delta = 0.34$.

Figure 7 shows the distributions of absorbed energy $Q(x, y)$ and their spectra for the laser pulses with the spiral intensity distribution obtained with OAMs having the topological charges $\ell = +1$, $+5$, and $+10$. In Fig. 7(a,b,c) we see that the spiral intensity pattern of the incident laser pulse is imprinted on the rough stainless steel surface in the form of large-scale spiral regions where absorption occurs. We can control the geometry of these spiral arrangements by changing the amplitude of the topological charge ℓ , which determines the number of spiral arms, and by its sign, which is responsible for the direction of the spiral twist. Within each arm of the spiral regions we see a chaotic distribution of absorbed energy spots aligned along the polarization direction, similar to that observed in the case of linearly

polarized laser pulses (see Fig. 2(a)). The spatial spectra of $Q(x, y)$ in Fig. 7(d,e,f) also resemble the spectrum obtained for a linearly polarized laser pulse with a flat phase (see Fig. 2(d)) with the difference that the large-scale spiral formations generate many spectral components at near-zero frequencies. Thus, we see that we can use OAM indirectly to create large-scale controllable arrangements of chiral LIPSS by generating spiral intensity distributions with given parameters.

IV. TWISTING OPTICAL FORCES

A. When orbital angular momenta do work

In the previous section we observed that despite our extensive efforts, we were unable to detect any direct effect of OAM on the distribution of absorbed laser energy during the interaction of laser pulses with rough surfaces. Therefore, there is a high probability of making a premature conclusion that OAM can not provoke the appearance of LIPSS with chirality. As we know from the literature, in certain situations, the laser pulses carrying OAM can sculpture chiral material structures [47, 60]. For ex-

ample, when a surface is irradiated by a nanosecond laser pulse with OAM, nanoscale twisted needles form in the region where the beam has the phase singularity. The twisting direction of these needles can be reversed by altering the sign of the topological charge ℓ . These observations have been made across various materials, including tantalum [45, 48], aluminum [61], copper [60], silicon [62], silver and gold thin films [46] and even azopolymers [63]. Interestingly, chiral surface relief formation was only observed when the handedness of the circular polarization aligned with that of the optical vortex. In contrast, it was suppressed when their signs were opposite [64]. This behavior highlights the effects of constructive and destructive coupling between SAM and OAM to achieve spiral surface reliefs. In these studies, optical radiation force has been widely invoked as the driving force for mass transport that occurs during the melting process.

B. Expression for optical forces

To understand in which cases laser pulses with OAM can transfer their vorticity to matter, let us consider the optical forces with which laser pulses act on the medium. In case of a single particle of charge q moving with velocity \vec{v} , the electromagnetic field of a laser pulse acts on this particle with the Lorentz force determined by the expression $q(\vec{E} + \vec{v} \times \vec{B})$. Therefore, if we have a material of volume V with the charge density ρ , then the overall force acting on this material from the electromagnetic field will be equal to $\int_V \rho(\vec{E} + \vec{v} \times \vec{B}) d^3r = \int_V (\rho\vec{E} + \vec{J} \times \vec{B}) d^3r$, where $\vec{J} = \rho\vec{v}$ is the current. According to this equation, the force \vec{f} acting on a unit volume of the material is given by $\vec{f} = \rho\vec{E} + \vec{J} \times \vec{B}$. Therefore, if we have a bulk material with an induced polarization \vec{P} , where the density of charges $\rho = -\vec{\nabla} \cdot \vec{P}$ and the current $\vec{J} = \partial\vec{P}/\partial t$, then the force applied to a unit volume of such material will be $\vec{f} = -(\vec{\nabla} \cdot \vec{P})\vec{E} + \partial\vec{P}/\partial t \times \vec{B}$. Considering a medium with a linear response, we can write $\vec{P} = \epsilon_0\chi\vec{E}$, where χ is the material susceptibility. Using this expression for the polarization \vec{P} together with the equality $\vec{B} = \mu_0\vec{H}$, we can finally express the force \vec{f} acting on a unit volume of the medium from the electromagnetic field of the laser pulse as

$$\vec{f} = -\epsilon_0\chi(\vec{\nabla} \cdot \vec{E})\vec{E} + \epsilon_0\mu_0\chi\frac{\partial\vec{E}}{\partial t} \times \vec{H}. \quad (5)$$

Here the first term, proportional to the gradient of the electric field, describes the component of the force pushing matter out of areas of high intensity. Assuming that longitudinal components of electric field are negligible, this component of the force acts in the transverse direction to the direction of laser pulse propagation. In turn, the second term describes the component of the force directed along the pulse propagation direction, parallel to the Poynting vector. In our studies, we focus primarily

on the intensity-dependent component of the force, as it is the only one capable of displacing matter parallel to the surface and thereby influencing the transverse layout of LIPSS. Another reason for our particular interest in the first term becomes evident when we consider the total force $\vec{f}_{\text{tot}} = \int_{-\infty}^{\infty} \vec{f}(x, y, z, t) dt$ which represents the accumulated force acting on the medium over time and continues to influence it even after the laser pulse has passed. According to Eq. (5) only the first term on the right-hand side contributes to \vec{f}_{tot} , since the time integral of the second term, containing the time derivative, is equal to zero. We can see this from the following simple considerations. Since the electric \vec{E} and magnetic \vec{H} fields oscillate at the same frequency, both of them can be described by the same harmonic function. In turn, the time derivative of \vec{E} results in a shift of half a period (e.g., the derivative of a sine is a cosine and vice versa). Therefore, $\partial\vec{E}/\partial t \times \vec{H}$ is an odd function of time whose temporal integral has to be zero. Thus, the total force \vec{f}_{tot} can be simply written as $\vec{f}_{\text{tot}} = -\epsilon_0\chi \int_{-\infty}^{\infty} (\vec{\nabla} \cdot \vec{E})\vec{E} dt$. In what follows we consider only the transverse optical forces given by the first intensity-dependent term in Eq. (5).

As a model laser pulse for studying optical forces, we consider a pulse whose electric field can be described by Eqs. (2) and (3) with the spatio-temporal amplitude $A(x, y, t)$ given by

$$A(x, y, t) = A_0 M(x, y) \sin^2\left(\frac{\pi}{2} \frac{t}{\tau_0}\right). \quad (6)$$

Similarly to Eq. (4), we assume that the temporal envelope is given by one period of \sin^2 function (for improved visual clarity, we use a shorter FWHM pulse duration $\tau_0 = 15$ fs). However, instead of a plane wave, here we consider a laser pulse with a beam shape determined by the function $M(x, y)$. In particular, we consider two beam shapes: a Gaussian beam defined by $M(x, y) = \exp(-r_\perp^2/2a_0^2)$ and a Laguerre-Gaussian beam, defined by $M(x, y) = (r_\perp/a_0)^{|\ell|} L_p^{|\ell|}(r_\perp^2/a_0^2) \exp(-r_\perp^2/2a_0^2)$, where $r_\perp = \sqrt{x^2 + y^2}$, $L_p^{|\ell|}$ are the generalized Laguerre polynomials with $p = 0$, and $a_0 = 10 \mu\text{m}$ is the beam radius. While the Gaussian beam serves as a good model for a generic laser beam, the Laguerre-Gaussian beam provides a more realistic description of laser pulses carrying the OAM with the topological charge ℓ [30].

C. Linear and circular polarizations

First, to give an intuitive idea of optical forces, let us consider a linearly polarized laser pulse ($\epsilon = 0$ and $\theta = 0^\circ$ in Eqs. (2) and (3)). Figure 8 shows the x components of the electric field, intensity, the snapshots of the transverse optical force at several points in time and the corresponding total force for the Gaussian and Laguerre-Gaussian beam shapes. The distributions of instantaneous optical force are presented at times $t = -T/4$,

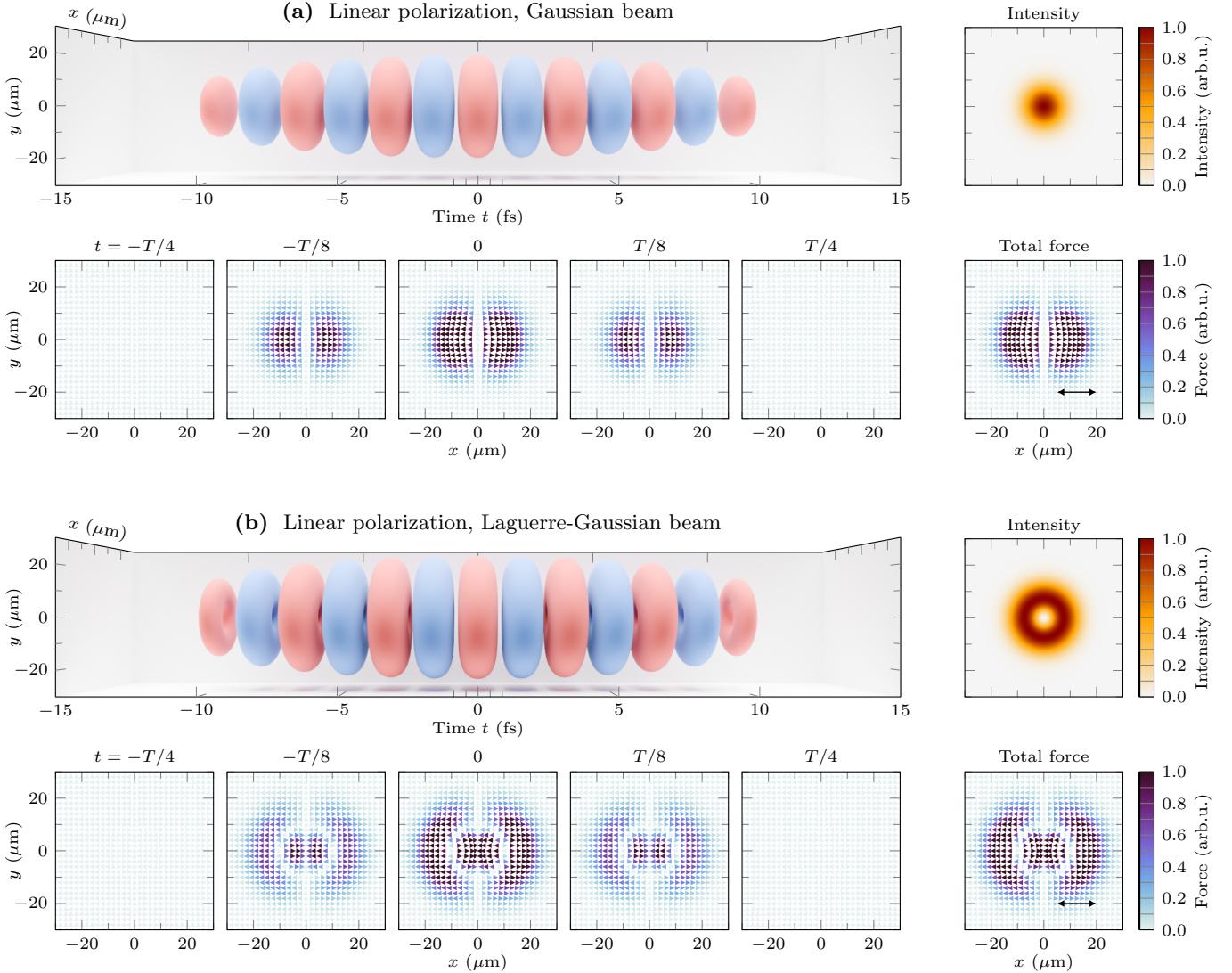


FIG. 8. Electric field E_x (isovalue at levels ± 0.1 V/m), intensity, the snapshots of optical force at times t within half an optical period T (the exact times are shown by minor ticks on the time axis) and the total force \vec{f}_{tot} for linearly polarized laser pulses with the Gaussian (a) and Laguerre-Gaussian (b) beam shapes. The arrows on the total force plot show the direction of laser polarization. The complete time evolution of optical forces is shown in the supplementary movies: `movie8a.mp4` for (a) and `movie8b.mp4` for (b).

$-T/8, 0, T/8, T/4$, that is within half the optical period $T = \lambda_0/c_0$. We visualize the force distributions only during one half of the period because, according to Eq. (5), the transverse optical force given by the first term depend on the intensity and thus have a periodicity of $T/2$. In Fig. 8 we see that the vectors of optical force are directed along the polarization direction, while their magnitude oscillates with the electric field, reaching its maximum at the field crests. We also observe that the optical force vectors, while remaining parallel to the polarization, point away from regions of high intensity. In particular, for the Laguerre-Gaussian beam, there are force components directed toward the dark core of the beam. Despite the oscillation of the force amplitude in time, the total force \vec{f}_{tot} for the both beam shapes mirrors the instantaneous force distributions.

Next, we consider the optical forces produced by circularly polarized laser pulses ($\epsilon = 1$ and $\theta = 0^\circ$ in Eqs. (2) and (3)). Figure 9 presents the x components of the electric field, intensity, the snapshots of the transverse optical force together with the total force for both beam shapes: Gaussian and Laguerre-Gaussian. Here we see that the amplitude of the optical force do not fluctuate with the field, but the force vectors, following the instantaneous direction of laser polarization, rotate along with the electric field vector. As a result of this rotation, the total force \vec{f}_{tot} becomes averaged over all possible angles. For the Gaussian beam shape, the vectors of \vec{f}_{tot} point in all directions from the beam center. In turn, for the

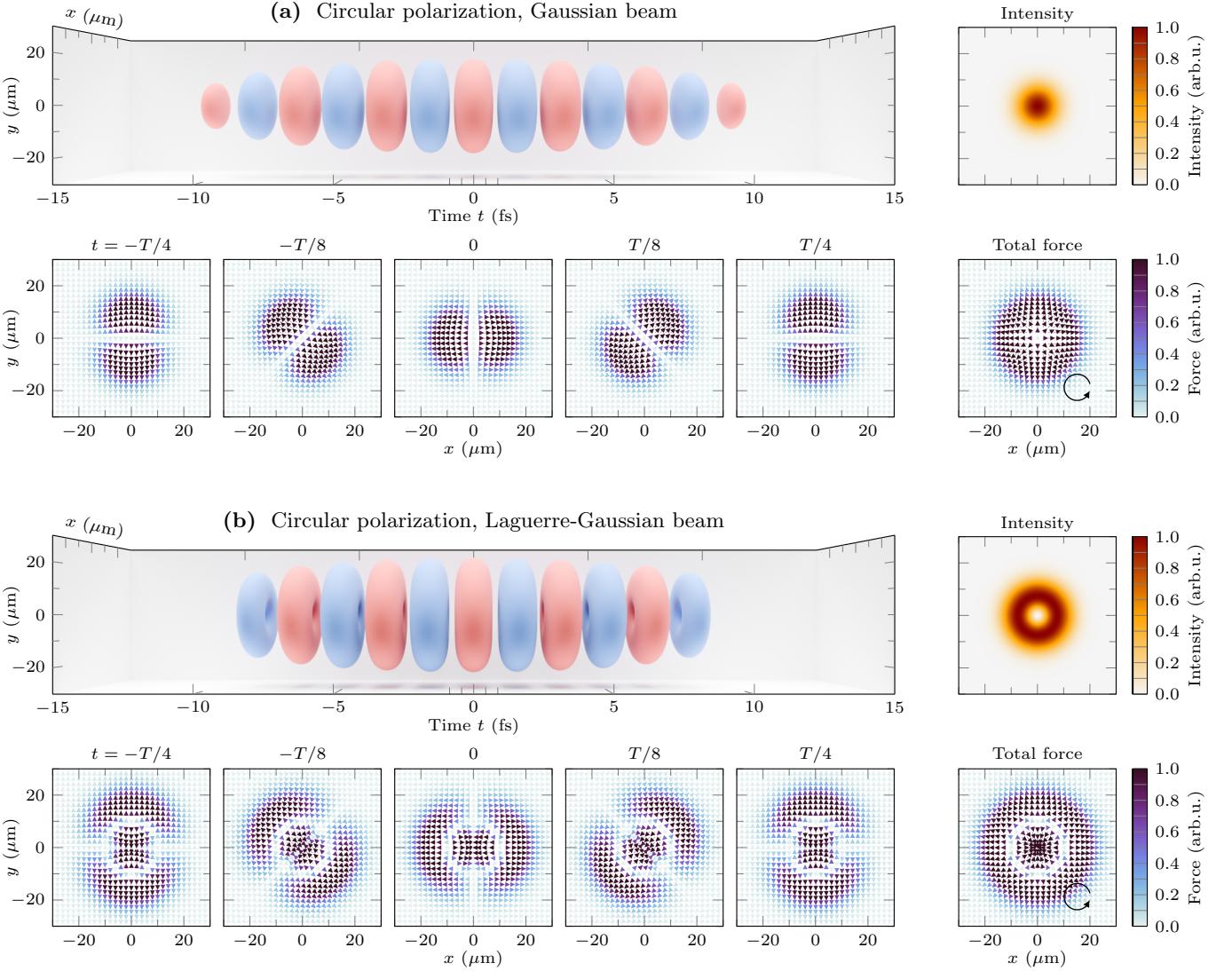


FIG. 9. Electric field E_x (isovalue at levels ± 0.1 V/m), intensity, the snapshots of optical force at times t within half an optical period T (the exact times are shown by minor ticks on the time axis) and the total force \vec{f}_{tot} for circularly polarized laser pulses with the Gaussian (a) and Laguerre-Gaussian (b) beam shapes. The arrows on the total force plot show the direction of laser polarization. The complete time evolution of optical forces is shown in the supplementary movies: `movie9a.mp4` for (a) and `movie9b.mp4` for (b).

Laguerre-Gaussian beam shape, the vectors of \vec{f}_{tot} , which also exhibit radially symmetric orientation, are directed both outward from the intensity ring and toward its center. In particular, we can speculate that the components of the total force directed inside the intensity ring lead to the formation of specific elevations observed in the center of the annular spot produced by Laguerre-Gaussian beams in experimental silicon nanostructuring [65].

D. Orbital angular momentum

To study the optical forces induced by OAM let us consider laser pulses with the Laguerre-Gaussian beam shape and the phase $\phi(x, y) = \ell \arctan(y/x)$. We examine three

different configurations of such laser pulses, defined using Eqs. (2) and (3): (i) a linearly polarized laser pulse with $\epsilon = 0$ and $\ell = +1$, (ii) a circularly polarized laser pulse with $\epsilon = 1$ and the same topological charge $\ell = +1$, and (iii) a circularly polarized laser pulse with $\epsilon = 1$ and the opposite topological charge $\ell = -1$. Figure 10 shows the x components of the electric field together with the laser pulse intensity, as well as the distributions of the optical force (both the time snapshots and the total force) for all three laser pulse configurations. In Fig. 10(a) we see that for the linearly polarized laser pulse the optical force vectors remain parallel to the polarization direction. However, the helical wavefront caused by OAM results in a distinctive rotation of the overall force distribution. Nevertheless, this rotation averages out over time, and

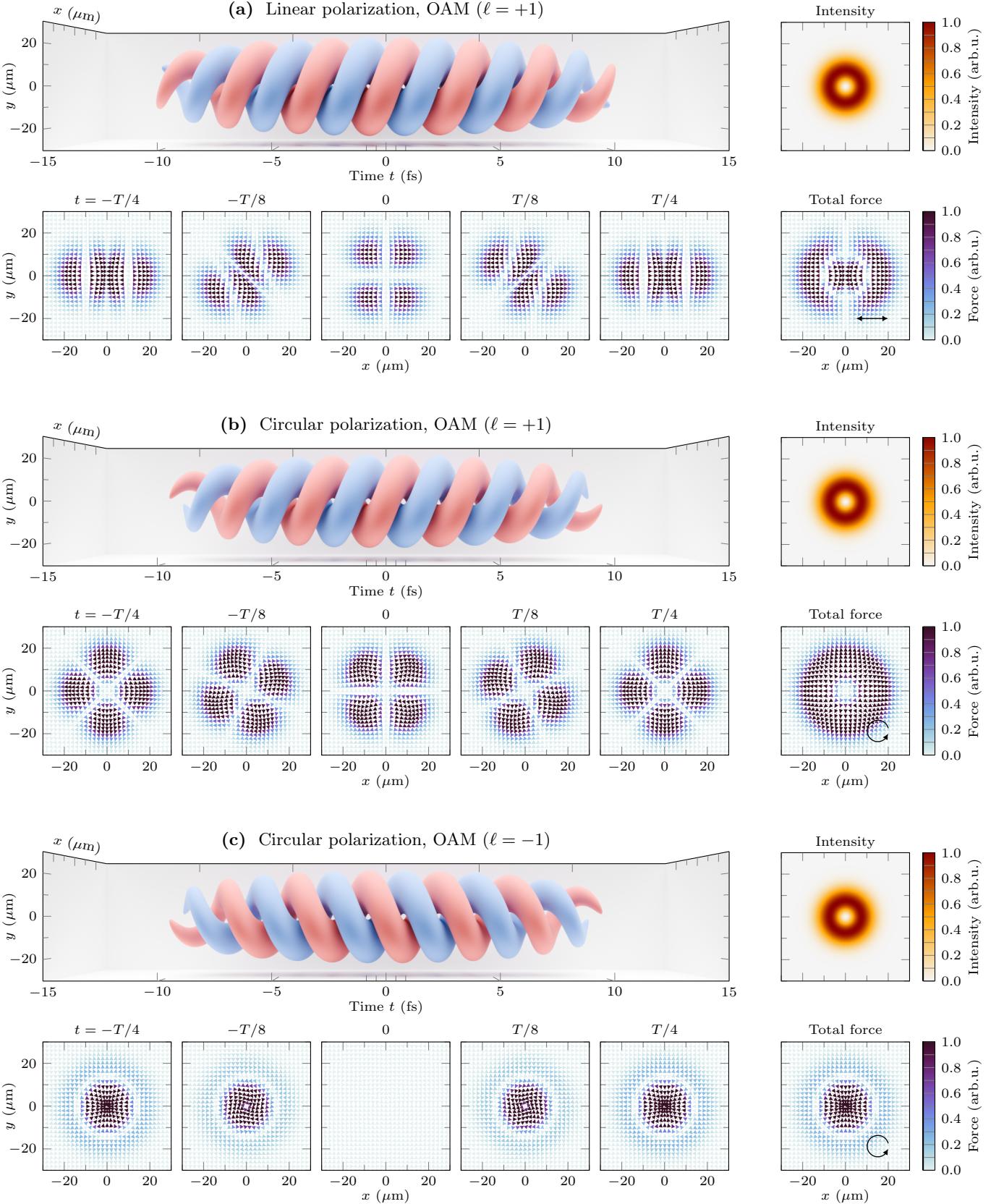


FIG. 10. Electric field E_x (isovalue at levels $\pm 0.1 \text{ V/m}$), intensity, the snapshots of optical force at times t within half an optical period T (the exact times are shown by minor ticks on the time axis) and the total force \vec{f}_{tot} for laser pulses carrying the OAM with the topological charge ℓ : (a) linear polarization with $\ell = +1$, (b) circular polarization with $\ell = +1$, (c) circular polarization with $\ell = -1$. The arrows on the total force plot show the direction of laser polarization. The complete time evolution of optical forces is shown in the supplementary movies: `movie10a.mp4` for (a), `movie10b.mp4` for (b), and `movie10c.mp4` for (c).

in the figure showing the total force distribution, there is no indication of the presence of OAM (compare with the total force distribution for a simple linearly polarized laser pulse in Fig. 8(b)).

Fig. 10(b) depicts the distributions of the optical force for a circularly polarized laser pulse, where the rotation of the polarization and wavefront occurs in the same direction ($\epsilon = 1$ and $\ell = +1$). As in the case of a circularly polarized laser pulse with a plane wave front (see Fig. 9(b)), we observe a comparable rotation of the optical force vectors, though with a different force distribution. In particular, we see that the force distribution has four distinct lobes. Also we note that there are no force components directed toward the dark core of the beam. As a result, the total force distribution consists only of force vectors directed outward from the beam center. We also note the absence of the net twisting force.

Finally, Fig. 10(c) shows the distribution of optical force for the circularly polarized laser pulse where the helical wavefront rotates in the direction opposite to the direction of laser polarization ($\epsilon = 1$ and $\ell = -1$). As we can see, such combination of the polarization rotation and the wavefront twist results in the force distribution without the lobes, in contrast to the case of co-rotating polarization and the wavefront shown in Fig. 10(b). This behavior of the force distribution reflects the results of adding and subtracting SAM and OAM. In Fig. 10(c) we also see that the force amplitude oscillates over time. In particular, we observe the appearance of the alternating twisting force (see the force snapshots at times $t = -T/8$ and $T/8$). However, because the twisting occurs in opposite directions, the resulting twisting averages out, causing all vectors in the total force distribution to align along the radial direction. Unlike the previous case of co-rotating polarization and wavefront (see Fig. 10(b)), here we see that most of the force is directed towards the beam center. As in the previous case, the distribution of \vec{f}_{tot} do not contain any net twisting force.

The above examples show that laser pulses with OAM are capable of twisting matter within the laser pulse duration even in the case of linear polarization. However, they do not leave any twisting force in the wake of the laser pulse as it passes. Therefore, to create chiral material structures with such laser pulses, the pulses need to be long enough for the pulse front to melt the material and the tail to induce a vortex. A multi-pulse configuration, where the first pulse melts the surface and subsequent pulses twist the molten material, could potentially achieve this effect as well.

E. Focused laser pulses

Above, we found that although laser pulses with OAM can introduce the twisting force, they exert the corresponding torque on the matter only during the pulse duration. However, the formation of chiral material structures would be much more efficient if the laser pulse could

create a net twisting force in its wake. In order to find such net twisting forces, let us consider focused OAM laser pulses. Note that in many LIPSS experiments the laser pulses are already focused on the sample surface, so adding external focusing in our analysis seems quite natural. Here we consider the same set of OAM laser pulses introduced previously (see Fig. 10): the linearly polarized laser pulse with $\epsilon = 0$ and $\ell = +1$, the circularly polarized laser pulse with $\epsilon = 1$ and $\ell = +1$ (co-rotating polarization and wavefront), and the circularly polarized laser pulse with $\epsilon = 1$ and $\ell = -1$ (counter-rotating polarization and wavefront). Similarly to the case of spiral intensity distribution, to model the external focusing we introduce the phase $\phi(x, y) = \ell \arctan(y/x) - k_0 \sqrt{x^2 + y^2} \sin \delta$, where the first term is responsible for the OAM and the second one for the focusing. For better visual appeal we consider a very smooth focusing with the convergence angle $\delta = 1^\circ$ corresponding to $NA = \sin \delta = 0.017$.

Figure 11 shows the x component of the electric field, laser pulse intensity, and distributions of optical force (instantaneous and total) for the focused OAM laser pulses introduced above. As we can see from the plots of the electric field, the presence of the phase term responsible for the focusing leads to a distortion of the wavefront: the electric field located closer to the beam center turns out to be lagging in time relative to the peripheral one. Note that the wavefront distortion affects the periodicity with which the optical force changes. Therefore, in the figures with the focused laser pulses, we plot the distributions of the instantaneous optical force at the moments of time $t = -T/2, -T/4, 0, T/4, T/2$, that is within the full optical period T , rather than its half, as we did for the previous figures. The figures with the instantaneous optical force show that the external focusing results in additional vorticity of the corresponding force distributions. Nevertheless, in Fig. 11(a) we see that despite this additional vorticity, the distribution of the total force \vec{f}_{tot} for a linearly polarized laser pulse does not indicate the presence of any net twisting force. However, in both cases of a circularly polarized laser pulse, we clearly see that the distributions of \vec{f}_{tot} have a residual vorticity. Thus, we can conclude that in focused laser pulses with OAM, it is the circular polarization that leads to the emergence of net twisting force. This effect is likely related to the recent observations of clockwise and counterclockwise nanopillar arrays fabricated using left- and right-handed circular polarizations at ZnO surface [66].

In light of the above observation, the question arises as to whether it is possible to obtain the net twisting force in focused circularly polarized laser pulses without OAM. To answer this question let us consider circularly polarized laser pulses with the Gaussian and Laguerre-Gaussian beam shapes focused by some external focusing element. We model such pulses by setting $\epsilon = 0$, $\theta = 0^\circ$ and phase $\phi(x, y) = -k_0 \sqrt{x^2 + y^2} \sin \delta$ in Eqs. (2) and (3) together with a corresponding beam shape in Eq. (6).

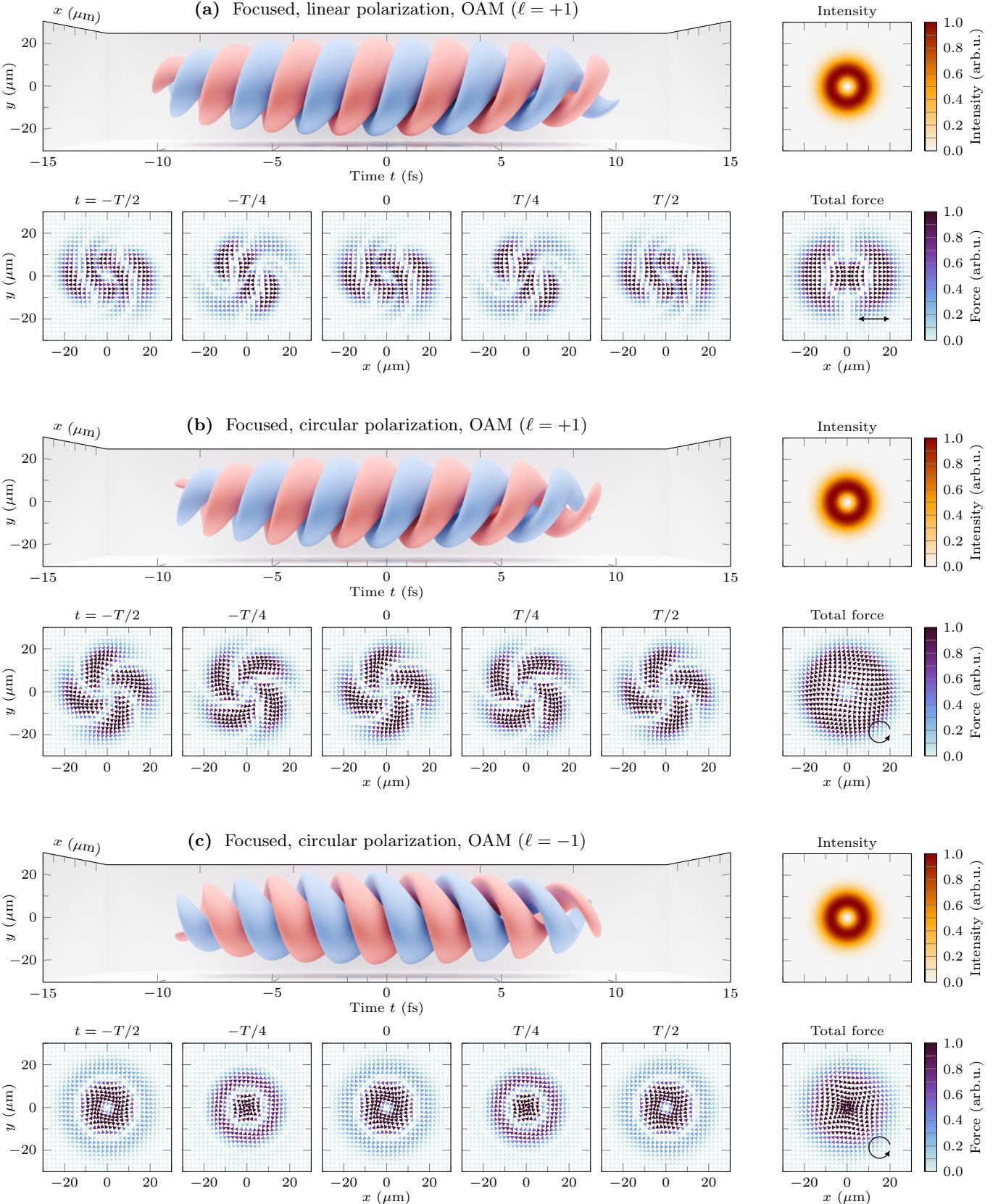


FIG. 11. Electric field E_x (isovalues at levels $\pm 0.1 \text{ V/m}$), intensity, the snapshots of optical force at times t within an optical period T (the exact times are shown by minor ticks on the time axis) and the total force \vec{f}_{tot} for focused laser pulses carrying an OAM with the topological charge ℓ : (a) linear polarization with $\ell = +1$, (b) circular polarization with $\ell = +1$, (c) circular polarization with $\ell = -1$. The arrows on the total force plot show the direction of laser polarization. The complete time evolution of optical forces is shown in the supplementary movies: `movie11a.mp4` for (a), `movie11b.mp4` for (b), and `movie11c.mp4` for (c).

Here we keep the same converging angle $\delta = 1^\circ$.

Figure 12 shows the electric field E_x , intensity, and distributions of optical force (both at different points in time and the total one) for focused linearly polarized laser pulses with the Gaussian and Laguerre-Gaussian beam shape. Compared to the case of unfocused circularly polarized laser pulses shown in Fig. 9, here we see that the optical force distributions form a spiral that rotates in time in the direction of laser polarization. In turn, on the plots of the total force we see that the vorticity of the instantaneous force distributions, accumulating over time, leads to a spiral arrangement of the force vectors, indicating the presence of a net twisting force. Thus, we can confirm that focused circularly polarized laser pulses can generate a twisting optical force that continues to act after the laser pulse has passed, potentially influencing subsequent thermo-mechanical processes [67].

V. CONCLUSIONS

Our results demonstrate the potential of vortex laser beams to induce unique surface morphologies on rough metallic surfaces. By varying polarization, orbital angular momentum, and initial pre-structures with chiral properties, we assess the conditions under which these beams can generate chiral excitations that result in intricate patterns, such as spiral and helical structures. Unlike conventional beams, the distinctive phase and polarization distributions of vortex beams enable the formation of complex, asymmetrical surface structures, providing new insights into the formation of LIPSS.

Contrary to our initial expectations, OAM in a laser pulse does not alter the distribution of absorbed laser energy on a rough surface or induce chiral LIPSS. Additionally, the presence of chiral structures does not ensure that the absorbed energy distribution will reflect OAM effects, making positive feedback from repeated pulses unlikely to enhance these effects. While we did not identify a configuration where OAM directly produces chiral distributions, we demonstrated that OAM can indirectly create a spiral intensity distribution by interfering with a plane wave. However, this approach does not achieve resolution below the laser wavelength. To explore mechanisms effective at the subwavelength scales, we examined the features of twisting optical forces.

Laser pulses with OAM exhibit varying behaviors based on polarization. For linearly polarized pulses with OAM, the optical force vectors align with the polarization but show a rotating distribution due to the helical wavefront, which averages out over time, leaving no net twisting force. Circularly polarized pulses with co-rotating OAM also show a rotation but lack force components directed toward the beam center, resulting in a radial force distribution. For counter-rotating OAM, the twisting force oscillates over time, leading to a net radial force with no residual twisting. Our investigation highlights that focused circularly polarized laser pulses,

with or without OAM, are crucial for generating a net twisting force, which could influence subsequent thermo-mechanical processes.

In conclusion, the combination of orbital and spin angular momenta significantly enhances the flexibility of surface functionalization. Structured light interacting with material surfaces enables advanced material processing with a level of control that surpasses conventional methods. As a key contribution of this work, we highlight the possibility of manipulating LIPSS via spiral intensity distributions and applying optical torque forces, expanding the applicability of LIPSS to areas such as biomimetic design, chiral sensing or enantiospecific surface physical chemistry. While we have shown that structured light can break conventional symmetry and induce rotating surface deformations, the development of self-formed coherent structures with adjustable chiroptical properties will open up new possibilities for diversifying the morphologies of LIPSS and designing advanced nanoarchitectures. To fully realize these potential applications, especially in nano-manufacturing, refining these methods is crucial. Further investigation should focus on how structured light interacts with more sophisticated materials, including those with inherent helical dichroism, to improve control over surface structures and enhance the functional applications of these chiroptical effects.

Appendix A: Phase-amplitude matching

If a laser pulse exhibits a non-uniform phase, its different spatial regions experience varying time delays. As a result, when modeling such a pulse, it is essential to account for these time delays in the field amplitude. To illustrate this, consider an x -polarized laser pulse with a central wavelength of $\lambda_0=1.03\text{ }\mu\text{m}$ and non-uniform phase $\phi(x, y)$ whose electric field can be represented by Eqs. (2) and (3) with $\epsilon = 0$ and $\theta = 0$. To clarify our arguments, we use a super-Gaussian amplitude in both space and time: $A(x, y, t) = \exp(-[\sqrt{x^2 + y^2}/a_0]^{10})\exp(-[(t - 2\tau_0)/\tau_0]^{10})$ with $a_0=10\text{ }\mu\text{m}$ and $\tau_0=10\text{ fs}$. According to Eq. (3a), the electric field of such laser pulse reaches its maxima when the argument of the cosine becomes zero, which occurs at moments of time

$$\tau = t + \phi(x, y)/\omega_0 = 0 \quad (\text{A1})$$

that determine the phase fronts. However, although the phase $\phi(x, y)$ changes the locations of the field maxima across the beam cross-section, it does not affect the temporal shape of the amplitude $A(x, y, t)$. If we ignore this fact, we risk obtaining an incorrect description of the electric field of the laser pulse. Let us demonstrate this with the following simple example. Imagine that we insert a transparent glass plate perpendicular to the direction of propagation of the laser pulse, such that only half of the laser beam passes through it. The part of the pulse passing through the glass plate will experience a

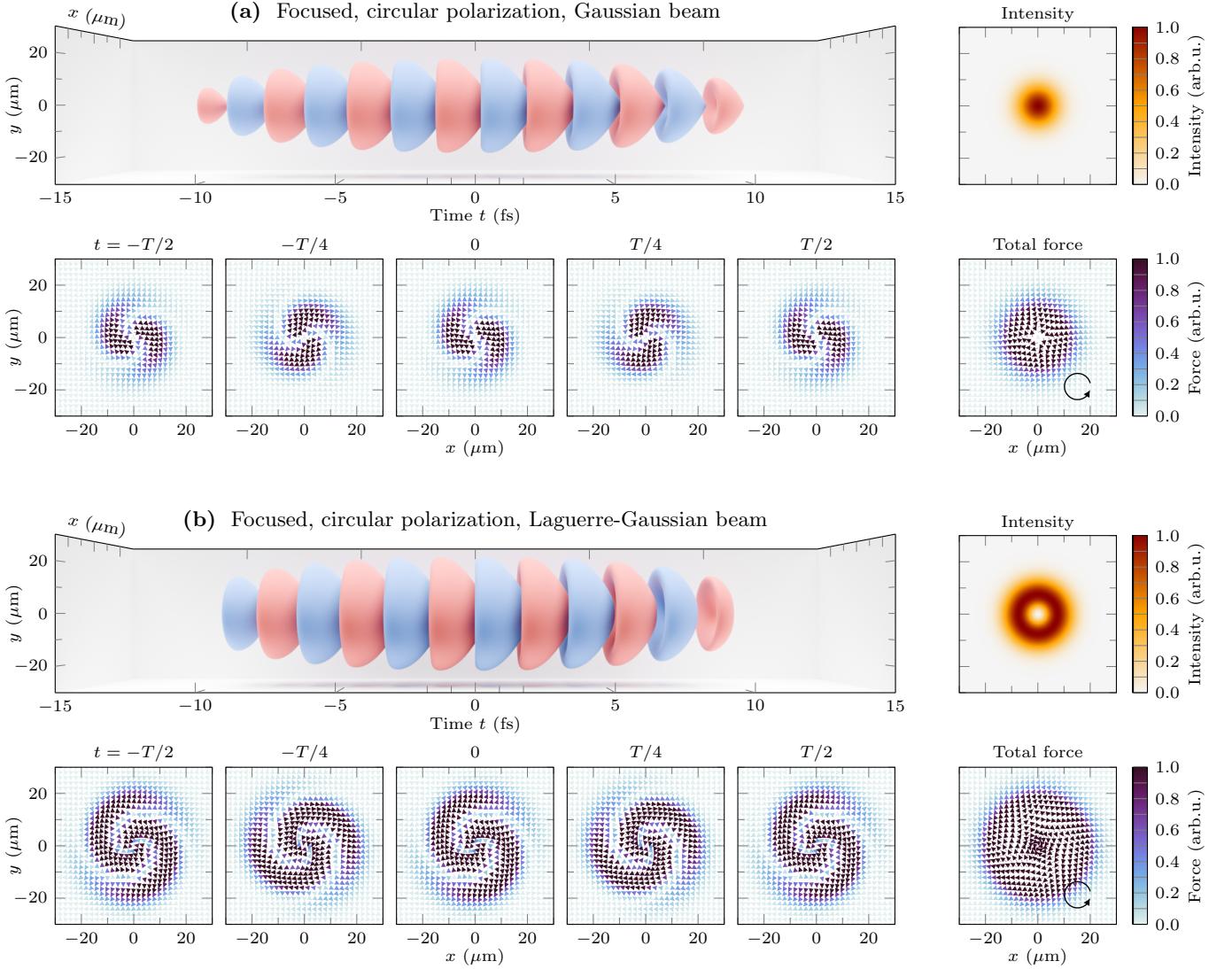


FIG. 12. Electric field E_x (isovalues at levels $\pm 0.1 \text{ V/m}$), intensity, the snapshots of optical force at times t within an optical period T (the exact times are shown by minor ticks on the time axis) and the total force \vec{f}_{tot} for focused circularly polarized laser pulses with the Gaussian (a) and Laguerre-Gaussian (b) beam shapes. The arrows on the total force plot show the direction of laser polarization. The complete time evolution of optical forces is shown in the supplementary movies: `movie12a.mp4` for (a) and `movie12b.mp4` for (b).

delay relative to the portion propagating in free space. We can model this situation by a step phase $\phi(x, y) = \varphi$ for $x < 0$ and zero otherwise, with the constant φ which depends on the thickness of the glass plate. As a result of such manipulations, the original laser pulse will be divided into two delayed in time parts, and, in an extreme case of a sufficiently thick glass plate (a sufficiently long delay), it will be split into two independent pulses. The original model of the laser pulse, given by the Eqs. (2) and (3), is not able to describe such a transformation of the laser pulse.

In Fig. 13(a) we plot the electric field and phase distribution for a specific case of a glass plate which introduces the phase delay $\varphi = 6\pi$. Here, we see that such a non-uniform phase does not result in any changes

to the electric field, which is clearly incorrect. This happens because according to Eq. (3a), $\varphi = 6\pi$ being a multiple of 2π , does not change the cosine part of electric field. To solve this problem, we apply the following time transformation to the electric field amplitude: $A(x, y, t) \rightarrow A(x, y, \tau)$, where $\tau = t + \phi(x, y)/\omega_0$. This transformation maps the phase front curvature (see Eq. (A1)) into actual time delays of amplitude. Figure 13(b) shows that the above time transformation of amplitude results in the part of the electric field, affected by the non-zero phase, being delayed in time. Thus, we see that this temporal transformation of amplitude enables us to model laser pulses with non-uniform phase distributions in accordance with physical reality.

As a second example, let us consider the effect of

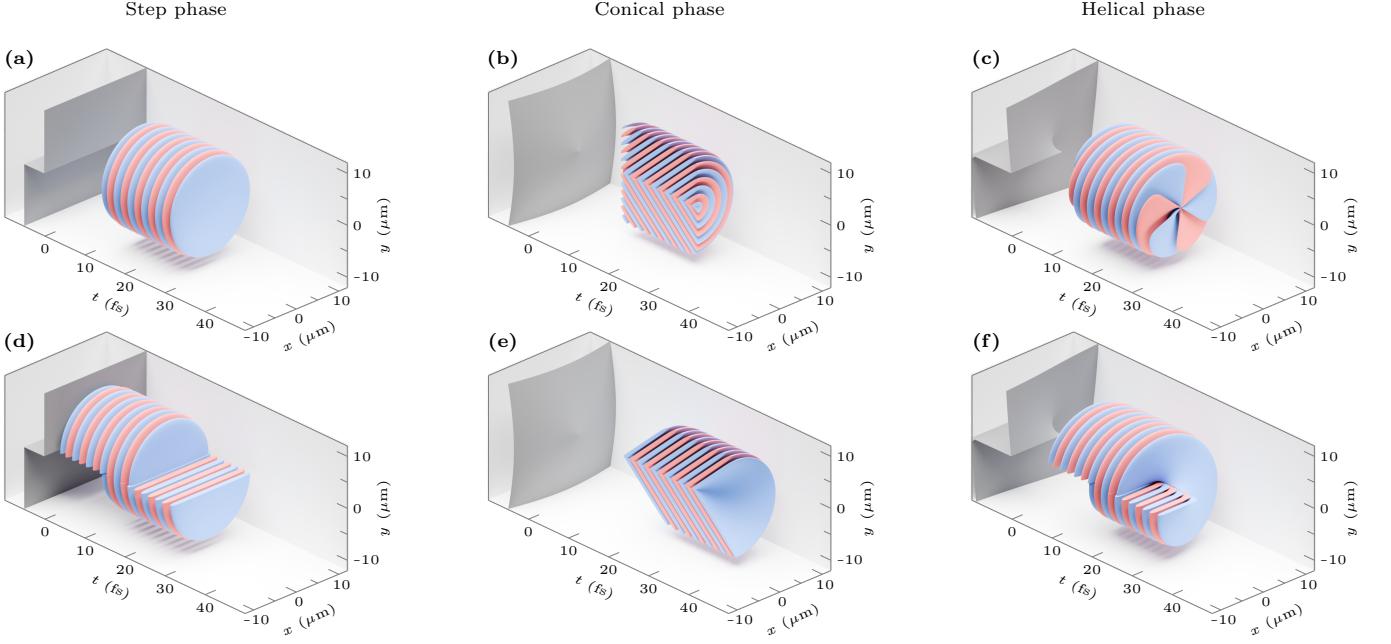


FIG. 13. Distributions of phase $\phi(x, y)$ (gray surfaces) and the corresponding electric fields (isosurfaces at levels ± 0.5 V/m) without (a)–(c) and with (d)–(f) time transformation of the amplitude. (a,d) Step phase, (b,e) conical phase, (c,f) helical phase. For the cone phase, the field isosurfaces are cut in half to better show the internal structure.

the conical phase $\phi(x, y) = -k_0\sqrt{x^2 + y^2} \sin(\alpha)$ which we use in the main text to model tightly focused laser pulses. Figures 13(c) and (d) show the distribution of phase $\phi(x, y)$ and the corresponding electric fields with and without the time transformation of the amplitude for the case of $\alpha=45^\circ$. In Fig. 13(c) we see that without the amplitude transformation the phase fronts have a conical structure, but the spatio-temporal shape of the laser pulse remains unchanged. This phase-amplitude mismatch will result in strong aberrations in the focal spot. However, as shown in Fig. 13(d), the amplitude transformation leads to a change in the time profile of laser pulse and its alignment with the conical phase fronts.

Finally, Figs. 13(e) and (f) show a more complex example demonstrating the effect of the amplitude transformation on a laser pulse with a helical wavefront given by the phase $\phi(x, y) = \ell \arctan(x/y)$ with $\ell = 3$. As Fig. 13(f) shows, the amplitude transformation allows us to correctly reproduce the rupture of the amplitude front along the line of the phase dislocation.

In summary, by using the time transformation of the

field amplitude given by $\tau = t + \phi(x, y)/\omega_0$, we can correctly describe the amplitude time delays caused by the inhomogeneous phase of $\phi(x, y)$. Although it is usually overlooked, proper phase-amplitude matching plays a key role in modeling of ultrashort pulses, especially in cases of tight focusing and strong near-field effects.

Keywords: structured light, orbital angular momentum, LIPSS

Data availability statement: The data generated during the current study are available from the authors upon reasonable request.

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