High-Harmonic Generation and Spin-Orbit Interaction of Light in a Relativistic Oscillating Window

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(Dated: October 5, 2020)

When a high power laser irradiates a small aperture on a solid foil target, the strong laser field drives surface plasma oscillation at the rim of the aperture, which acts as a “relativistic oscillating window”. The diffracted light travels though such an aperture contains high-harmonics of the fundamental laser frequency. When the driving laser is circularly polarised, the high-harmonic generation (HHG) process facilitates a conversion of the spin angular momentum of the fundamental light into the intrinsic orbital angular momentum of the harmonics. By means of theoretical modelling and fully 3D particle-in-cell simulations, it is shown the harmonic beams of order \( n \) are optical vortices with topological charge \( |l| = n - 1 \). When the driving laser is significantly intense, the HHG is dramatically enhanced through a surface wave breaking effect, which leads to a universal power-law spectrum \( I_n \sim n^{-3.5} \) at ultra-relativistic limit, where \( I_n \) is the intensity of the \( n \)th harmonic. This work opens up a new realm of possibilities for producing intense extreme ultraviolet vortices, and diffraction-based HHG studies at relativistic intensities.

Light carries angular momentum as spin and orbital components. The spin angular momentum (SAM) is associated with right or left circular polarisation (±\( \hbar \) per photon), and the orbital angular momentum (OAM) is carried by light beams with helical phase fronts exp(\( il\phi \)) (\( \hbar \) per photon), also known as optical vortices, where \( l \) is the topological charge and \( \phi \) is the azimuthal angle [1]. The spin-orbit interaction of light refers to phenomena in which the spin affects the orbital degrees of freedom [2], such as spin-Hall effects [3, 4], spin-dependent effects in nonparaxial fields [5] and evanescent waves [6]. Recently, interest in spin-orbit interaction has surged, as it provides physical insight into the behaviour of polarised light at sub-wavelength scales, which is essential in nano-optics and photonics. In addition, spin-orbit angular momentum conservation is also an important concept to produce optical vortices [11], that have rich variety of applications in optical communication [7, 8], biophotonics [9], and optical trapping [10].

The production of optical vortices with such methods mostly rely on high-harmonic generation (HHG) in laser-atom interactions, driven by a moderately intense (\( \sim 10^{14} \text{W/cm}^2 \)) beam [11–15]. The resulting extreme ultraviolet (XUV) vortices are of particular interest for monitoring and manipulating the SAM and OAM of light-matter interactions on the atomic scale, as well as for applications such as nonlinear optics [16, 17] and superresolution microscopy [18]. Owing to the remarkable progresses in high-power lasers [19], such advanced light sources open up new possibilities in the relativistic regime (\( n > 10^{18} \text{W/cm}^2 \)) of laser-matter interactions [20–25], and can yield fundamental insights into the spin-orbit/orbit-orbit angular momentum interactions at ultra-high intensities [26–29]. It is reported recently [30] that by irradiating a solid foil with a circularly polarised (CP) high-power laser, the SAM of the driver can be converted into OAM of the harmonic beams through the relativistic oscillating mirror (ROM) mechanism [31–33], giving rise to intense, ultrafast XUV vortices. However, according to the ROM theory [33], HHG is suppressed for CP driver at normal incidence. Therefore the mechanism relies crucially on the pre-denting of the target surface by radiation pressure, and typically produces relatively weak harmonic intensities [30].

Here we present, for the first time, a semi-analytical theory of HHG based on light diffraction at relativistic intensities. A new HHG mechanism is identified, which we call relativistic oscillating window (ROW). It allows for producing harmonic beams efficiently with a CP driving laser, and simultaneously, facilitating spin-to-orbital angular momentum conversion that produces ultra-intense XUV optical vortices. The ROW mechanism relies on a high-contrast [34] relativistic laser travelling through a small aperture on a thin foil, with dynamic surface electron oscillation on the rim, driven by the strong laser field. We demonstrate that the diffracted light through such a window contains both even and odd harmonics of the fundamental driving laser frequency; a universal power-law spectrum \( I_n \sim n^{-3.5} \) is produced for any sufficiently intense CP driver, where \( n \) is the harmonic order and \( I_n \) is the intensity of \( n \)th harmonic; all the harmonic components (\( n \geq 2 \)) are optical vortices with topological charge \( l = (n - 1)\sigma \), where \( \sigma \) denotes the right (\( \sigma = +1 \)) or left (\( \sigma = -1 \)) handed circular polarisation of the driving laser.

Results

We first demonstrate our scheme with 3D particle-in-cell (PIC) simulations. The simulation setup and the main results are summarised in Fig. 1: a right-handed CP laser irradiates normally on a solid foil located at...
FIG. 1: Generation of high-harmonic optical vortices in a ROW. (a) A circularly-polarised high-power laser is focused on a foil target with a small aperture, the intense laser fields drives surface electron oscillation on the rim of the aperture resulting in a dynamical electron density distribution (b). The three snapshots are separated temporally by a third of laser period ($T_0$), from left to right, and the white dashed lines represent the boundary of a rigid oscillating window. (c) The spectrum of the diffracted light through such an oscillating window, the red dashed line represents a fitted power-law spectrum $I_n \propto n^{-3.5}$. (d-f) show the 3D structures of the harmonics with frequency $2\omega_0$, $3\omega_0$, and $4\omega_0$, respectively. The field distribution in the plane marked by dark green colour in (d-f) are shown in (g-i), respectively.

$x_0 = 4 \mu\text{m}$ with a small aperture (radius $r_0 = 3.3\mu\text{m}$) aligned with the laser beam [see Fig. 1(a)], the thickness of the foil is $L_f = 0.25\mu\text{m}$. The intensity of the laser is $I_0 \approx 6.8 \times 10^{19} \text{ W cm}^{-2}$, corresponding to a normalised laser amplitude of $a_0 = eE_0/m_e c \omega_0 = 5$, where $E_0$ is the laser field, $e, m_e, c$ and $\omega_0$ denote the elementary charge, electron mass, vacuum light speed, and the laser frequency, respectively. The simulation parameters are detailed in Methods. With such intensity, the laser drives strong surface electron oscillations on the rim of the aperture, which modifies the local plasma density as shown in Fig. 1(b). The region with electron density above the critical density ($n_c = n_0 \omega_0^2/4\pi e^2 \approx 1.1 \times 10^{21} \text{ cm}^{-3}$) is reflective to the laser, therefore the transparent area [indicated by the white dashed lines in Fig. 1(b)], where light can travel through, acts as a “relativistic oscillating window”.

The diffraction of light through such an aperture generates high-harmonics of the fundamental driving laser frequency. Figure 1(c) presents a typical HHG spectrum; one can see that both even and odd harmonic orders are generated, and the spectrum has a power-law shape that can be fitted by $I_n \propto n^{-3.5}$. The spectrum is obtained as Fourier transform of the fields recorded at an observational plane $11 \mu\text{m}$ away from the screen ($x = x_0 + 11 \mu\text{m}$), and is averaged within an opening angle of $\theta = 30^\circ$.

Each harmonic with order $n$ is then selected by spectral filtering in the frequency range $[n-0.5,n+0.5]\omega_0$, as shown by Fig. 1(d-i). Notably, the spin-orbit interaction of light takes place in the HHG, all harmonics ($n \geq 2$) are optical vortices with topological charge $l = (n-1)\sigma$. The sign of $l$ is controlled by the polarisation of the driving laser ($\sigma$), which determines the chirality of the oscillating window, and has a profound impact on the orbital degree of freedom of the harmonics.

To understand these results, one must consider the diffraction of light through an oscillating aperture on a screen, for which the boundary condition is crucial. As discussed by Baeva et al. [33], the tangential component of electric field associated with a plasma surface current is negligibly small compared to the laser field $E_0$. Therefore it is reasonable to assume that on the
rear side of the screen, the tangential components of
electric field vanish everywhere except in the aperture,
where they can be approximated by that of the incoming
laser fields. For the sake of simplicity, we will restrict
our attention to the case of a monochromatic plane wave
\( \mathbf{E}(x, y, z, t) = \mathbf{U}(x, y, z) \exp(-i\omega_0t) \) normally incident on
the screen, where \( \mathbf{U}(x, y, z) \) satisfies Helmholtz equation
\( (\nabla^2 + k_0^2)\mathbf{U} = 0 \), and \( k_0 = \omega_0/c \) is the laser wavenumber.
For a stationary aperture, the solution is given by the
generalised Kirchhoff integral [35]:
\[
\mathbf{U}(x, y, z) = \frac{1}{2\pi} \nabla \times \int_A (\mathbf{e}_n \times \mathbf{U}) \exp\left(\frac{i k_0 R}{R}\right) ds',
\]
where the integration is only over the aperture on the
screen, \( \mathbf{e}_n \) is the unit vector rightward directed normal
to the screen, \( R = \sqrt{(x-x')^2 + (y-y')^2 + (z-z')^2} \) is the distance from the elementary source \( ds' \)
at \((x', y', z')\) to the observation point \((x, y, z)\).

For the situation under consideration in this work, each
\( ds' \) can be considered to be shaken by the laser field,
and \( R \) becomes time-dependent, leading to nonlinear
effects. We now introduce the ROW model, it assumes
that the shape of the aperture does not change (rigid win-
dow), such that each element \( ds' \) in the integral Eq. (1) is
shifted by the same amount of displacement \( d\mathbf{R}(t) \). This
assumption is valid for weakly-relativistic drivers (laser
intensity \( a_0 \leq 0.3 \)), we will extend the model to ultra-
relativistic intensity \( (a_0 \gg 1) \) later. The diffracted field
is then given by
\[
E_{\text{diff}}(x, y, z, t) = \mathbf{U}(x, y, z) \exp(-i\omega_0t)
= \frac{1}{2\pi} \nabla \times \int_A (\mathbf{e}_n \times \mathbf{U}) \exp\left[\frac{i k_0 R'(t') - i\omega_0t}{R'(t')}\right] ds',
\]
where \( R'(t') = |\mathbf{R} - d\mathbf{R}(t')| \) is the distance measured at
retarded time \( t' = t - R'(t')/c \).

Due to the rigid window assumption, \( R'(t') \) can be
obtained from the electron dynamics on the boundary
of the aperture, which depends on the details of laser-
plasma interaction. Note that Eq. (2) is valid for both
circularly and linearly polarised (LP) drivers, however
the respective behaviours of \( R'(t') \) are distinct. For a LP
driver, the window oscillates mostly in the polarisation
direction, which produces HHG beams carrying no OAM.

For the purpose of the present work, we will proceed
with CP drivers. At weakly-relativistic intensities,
the surface electrons are simply shifted antiparallel to the
driving laser field, resulting in a harmonic oscillation
\( d\mathbf{R}(t') = -i(\mathbf{e}_y + i\sigma \mathbf{e}_z)\delta r_0 \exp(-i\omega_0t') \), where \( \mathbf{e}_y \ (\mathbf{e}_z) \)
is the unit vectors in \((y \ (z))\) direction, and the amplitude
\( \delta r_0 \) is limited to \( \delta r_0 \leq k_0^{-1} \) in this case. The rim of the
window is always attached to these oscillating electrons.
To calculate the diffracted fields, one must solve for the
retarded time \( t' = t - R'(t')/c \) numerically according to
the motion of the source:
\[
R'(t') = |\mathbf{R} + (\mathbf{e}_y + i\sigma \mathbf{e}_z)\delta r_0 \exp(i k_0 R'(t') - i\omega_0t')|.
\]

However, to explain the spin-orbit interaction in the
HHG process, it is sufficient to derive analytically
the lowest order of diffracted fields, valid for \( a_0 \ll 1 \), seen
by a distant, paraxial observer, that satisfies \( (R \gg
r = \sqrt{y^2 + z^2} \gg r_0, \delta r_0) \). In this case we have
\( R'(t') \approx R + \delta r_0 \sin(\theta) \exp(ik_0 R' - i\omega_0t + i\sigma \phi) \),
where \( \theta = \arctan[r/(x-x_0)] \) is the diffraction opening an-
gle, and the azimuthal angle \( \phi \) is measured counterclockwise with respect to the \( y \)-axis in the \( y-z \) plane
[see Fig. 1(a)]. The phase term in Eq. (2) is then
\( \Phi = k_0 R'(t') - \omega_0 t_0 + c \exp(i k_0 R' - i\omega_0 t + i\sigma \phi) \),
where \( \epsilon = k_0 \delta r_0 \sin(\theta) \ll 1 \). Substituting it into Eq. (2)
and make use of the Jacobi-Anger identity [36], yield:
\[
E_{\text{diff}} \approx E_0(\mathbf{e}_y + i\sigma \mathbf{e}_z) \sum_{n=1}^{\infty} \frac{-i\epsilon R_0}{2\pi n \sin(\theta)} J_{n-1}(\epsilon n \sigma) \exp\left[\frac{in k_0 R_0 - \omega_0 t_0 + i(n-1)\sigma \phi}{R_0}\right],
\]
where \( R_0 = \sqrt{(x-x_0)^2 + y^2 + z^2} \) is the distance mea-
sured from the centre of the aperture, and \( J_n \) are the
Bessel functions of the first kind.

Equation (4) shows that HHG beams have the same
similar circular polarisation state as the driving laser, and their
phase fronts are all helical, with a topological charge
\( l = (n-1)\sigma \) for the \( n \)th harmonic. It agrees well with
the main findings from PIC simulations shown in Fig. 1.
From a quantum optics point of view, this relation guar-
gurantees the conservation of total angular momentum
and energy: when \( n \) photons at the fundamental frequency
are transformed into one photon of \( n \)th-order harmonic,
their SAM \((n\sigma)\) are converted into \((n-1)\sigma\) OAM
plus \( \sigma \) SAM.

We wish to examine the ROW model in detail for
drivers with higher intensity, by comparing the HHG
spectra and the diffracted field distribution obtained
from the model and the PIC simulations. For this pur-
pose, Eqs. (2-3) are solved iteratively [32]. Figure 2(a)
presents a typical solution of Eq. (3) for the distance be-
tween the centre of the aperture and an observer located
11 \( \mu \)m away from the screen with \( \theta = 30^\circ \). It shows
that due to the time it take for the light to travel from
the source to the observer, a harmonic oscillation of the
source results in an anharmonic oscillation seen by the
observer. This distortion due to retardation is the dom-
inant mechanism to generate the high-harmonics.

Figure 2(b) shows the HHG spectra obtained from PIC
simulations for weakly-relativistic drivers. The intensi-
ties of HHG beams increase dramatically with laser \( a_0 \).
In particular, the spectra for small \( a_0 \) decays faster than
exponentially with harmonic order \( n \), only the second-
order harmonic is visible for \( a_0 = 0.1 \). This agrees with
Eq. (4) as \( J_{n-1}(\epsilon) \sim (\epsilon/2)^{n-1}/(n-1)! \). As \( a_0 \) grows,
the spectra asymptotically converges to a power-law shape
\( I_n \propto n^\beta \). This trend can be reproduced by our model as
indicated by the open circles in Fig. 2(b).
However, the model suggests that the power-law exponent $\alpha$ is limited to around $\alpha = -8.7$, since the amplitude $\delta r_0$ should be smaller than $k_0^{-1}$. According to Fig. 2(b), this is only true for $a_0 \leq 0.3$, because at higher intensities, the electrons that oscillate on the boundary of the aperture may gain enough energy to escape the restoring force of plasma [37–39], as shown by the inset in Fig. 2(c). Therefore, the rim of the window can no longer be considered to be attached to these electrons, which significantly modifies the dynamics of the ROW.

This effect is essentially surface wave breaking (SWB) [40], that occurs when the driving force is too strong. To quantify when it should be taken into account, in Fig. 2(c) we plot the ratio of electron number that is emitted from the rim of the aperture divided by the total electron number in the skin layer, plotted against the laser amplitude $a_0$, the inset in (c) shows a typical electron density distribution in $x$-$y$ plane, when surface wave breaking occurs.

Figure 3(a) presents a snapshot of typical plasma density distribution near the aperture when SWB occurs. The shape of the window, bounded by the solid curves (red and green), is no longer circular. The SWB effect allows the electrons to excurse far into the aperture, as they travel inwards. The rim of the window on this side (red solid curve), follows the motion of the electrons for about half of one laser cycle, where the maximum displacement is $\sim c \times 0.5T_0 = 0.5\lambda_0$. Afterwards, most of these electrons are emitted away, and the rim of the window falls back to the original boundary as the local plasma density drops and transparency is restored. On the other side (green solid curve), when the electrons travel towards the plasma bulk, the displacement remains small ($\sim k_0^{-1}$).

The diffracted field through such a deformed oscillating window can be calculated by separating the aperture into two parts, A1 and A2. As shown by Fig. 3(a), they are fractions of two rigid windows (represented by the red and green circles, consisting of both solid and dashed lines), which oscillate with different amplitudes $\delta r_{A1} > \delta r_{A2} \approx k_0^{-1}$. The contributions from each part can then be obtained by integrating Eq. (2) over the area that satisfies $\mathbf{r} \cdot \mathbf{dR}'(t') \leq 0$ and $\mathbf{r} \cdot \mathbf{dR}'(t') > 0$ for A1 and A2, respectively.

In this way, the ROW model can be extended to ultra-relativistic intensities as shown by Fig. 3(b). One can see the exponent of power-law spectrum is increased dramatically by the SWB effect, which allows half of the window to oscillate with a larger amplitude. The HHG spectra for $a_0 > 0.3$ can be reproduced from the model by adjusting the value of $\delta r_{A1}$. Setting $\delta r_{A1} = 0.25\lambda_0$ and $0.4\lambda_0$ recovers the HHG spectra from PIC simulations with $a_0 = 0.5$ and 1 (adjusted to the fifth harmonic), respectively.

In particular, comparing the HHG spectra for $a_0 = 2$, $a_0 = 10$ [black and red colours in Fig. 3(b)], and $a_0 = 5$ [Fig. 1(c)], one can infer that the power-law exponent is limited to around $\alpha = -3.5$. This can be easily understood from our model, as the maximum displacement of electron layer is $0.5\lambda_0$. Thus, setting $\delta r_{A1} = 0.5\lambda_0$ gives a universal HHG spectrum for any sufficiently strong CP drivers. The results are presented by the black open circles in Fig. 3(b), which agree very well with the PIC simulations. In addition, both the ROW model and PIC

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**FIG. 2: ROW model in the harmonic oscillation regime for weakly-relativistic drivers.** (a) The temporal evolution of the distance between the ROW centre and an observer. The harmonic oscillation of the ROW is presented by the black curve, and the motion seen by the observer, obtained by solving Eq. (3), are shown with the red and blue lines, representing the results after one and two iterations, respectively. (b) HHG spectra for weakly-relativistic drivers, the green, blue, black, and red solid lines shows the results from PIC simulations with $a_0 = 0.1, 0.2, 0.3$, and 0.4, respectively. The black open circles show the prediction of ROW model in the harmonic oscillating case, with $\delta r_0 = k_0^{-1}$, and the black dashed line shows the fitting by $I_n \propto n^{-8.7}$. (c) The number of electrons emitted from the rim of the aperture divided by the total electron number in the skin layer, plotted against the laser amplitude $a_0$, the inset shows a typical electron density distribution in $x$-$y$ plane.
FIG. 3: **ROW model for ultra-relativistic drivers – the surface wave breaking effect.** (a) The electron density distribution near the aperture driven by a laser with amplitude $a_0 = 2$. The window (transparent area) is bounded by the solid curves (red and green). It can be separated into A1 and A2, which are fractions of two other rigid ROWs (red and green circles, consisting of both solid and dashed lines), with amplitude $\delta r_{A1}$ and $\delta r_{A2}$, respectively. (b) The HHG spectra obtained from PIC simulations ($r_0 = 3.3\lambda_0$ is fixed) with $a_0 = 0.5$, 1, 2, and 10 are presented by the green, blue, black, and red solid lines, respectively. The black open circles shows the prediction of ROW model taken into account the SWB effect, with $\delta r_{A1} = 0.25\lambda_0$ (green), 0.4\lambda_0 (blue), and 0.5\lambda_0 (black), respectively. (c) The HHG spectra with different aperture radii $r_0 = 2.3\mu$m and 4.3\mu m, the PIC simulation data ($a_0 = 5$ is fixed) are shown by the blue and red lines, while the results from the ROW model ($\delta r_{A1} = 0.5\lambda_0$) are presented by the blue open circle and red crosses, respectively. The $E_y$ field distribution of the (d) second, (e) third, and (f) fourth harmonics obtained from the ROW model.

Simulations suggest this limit change little with varying the radius of the aperture, two examples are given in Fig. 3(c).

Finally, the HHG fields can be obtained by filtering the diffracted fields from the ROW model within a certain frequency range. Using the same parameters as in Fig. 1, and setting $\delta r_{A1} = 0.5\lambda_0$, the corresponding second, third, and fourth harmonics are presented in Fig. 3(d-f), respectively. Apparently the results confirm the relation $l = (n - 1)\sigma$, and the harmonic field distributions agree very well with the PIC simulations shown in Fig. 1(g-i).

In conclusion, we have demonstrated that high-harmonic beams are generated due to the diffraction of a high-power CP laser through a small aperture on a solid plasma foil. In this process, the spin angular momentum of the driving laser is converted into orbital angular momentum of the harmonics, giving rise to intense optical vortices in the XUV regime. Three dimensional PIC simulations show that a power-law high-harmonic spectrum $I_n \propto n^\alpha$ is produced, and a universal exponent $\alpha \approx -3.5$ is observed for sufficiently strong ($a_0 \geq 2$) CP drivers. The topological charge of the $n$th harmonic light is $l = (n - 1)\sigma$. The high-harmonic generation and spin-orbit interaction of light stem from the chiral electron oscillation on the rim of the aperture, that act as a “relativistic oscillating window”. Based on this picture, a semi-analytical model is developed, which agrees well with the numerical findings.

**Methods**

**Laser-plasma parameters.** In the simulation presented in Fig. 1, a circularly-polarised high-power laser beam enters the simulation box from the left ($-x$) boundary and propagates to the right. The laser field is $E_0 = (e_y + i\sigma e_z)E_0\sin^2(\pi t/\tau_0)\exp(ik_0x - i\omega_0t)$, where $0 < t < \tau_0 = 54$ fs, $E_0 = 16$ TV m$^{-1}$ is the laser amplitude, frequency $\omega_0 = k_0c$, wavenumber $k_0 = 2\pi/\lambda_0$ with
$\lambda_0 = 1 \mu m$ the laser wavelength. The laser polarisation state is controlled by $\sigma = +1$ for RCP and $-1$ for LCP. In Figs. 2-3, the intensity of laser is changed while other parameters are kept the same.

Note that we have assumed that the laser focus spot is much greater than the size of the aperture, so that the intensity on the edge of the aperture does not depend on the its radius. This is only for the convenience of comparing the PIC results with our model, not crucial for the proposed mechanism to work.

The foil target [assumed plastic (CH)] is modelled by a pre-ionised plasma with electron density $n_0 = 30n_c$, thickness $L_t = 0.25 \mu m$. The radius of the aperture are $r_A = 4.0 \mu m$. In order to account for heating by the laser pre-pulse, the inner boundary ($r < r_A$) is assumed to have a density gradient $n(r) = n_0 \exp[(r - r_A)/h]$, with scale length $h = 0.2\mu m$, which yields an effective radius $r_0 = 3.3\mu m$, for which $n(r_0) = 1n_c$.

**PIC simulations.** The 3D PIC simulations presented in this work were conducted with the code EPOCH [41], and the algorithm developed by Cowan et al. [42] is used to minimise the numerical dispersion. For most of the simulations presented in this paper, the dimensions of the simulation box are $x \times y \times z = 15\mu m \times 16\mu m \times 16\mu m$, sampled by $2400 \times 320 \times 320$ cells with four macro particles for electrons, two for C$^{6+}$ and two for H$^+$. Ions. A higher transverse resolution $x \times y \times z = 10\mu m \times 10\mu m \times 10\mu m$, sampled by $1000 \times 500 \times 500$ cells, and 14 macro electrons per cell is used to simulate the fine details of density fluctuation in the ultra-relativistic case, presented in Fig. 3(a). A high-order particle shape function (fifth order particle weighting) is applied to suppress numerical self-heating instabilities (see Sec. 5.1 of ref.[41] for details).

**Data availability.**

The data that support the findings of this study are available from the corresponding author upon request.

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Acknowledgements

The author acknowledges fruitful discussions with A. Pukhov, T. Fülöp and I. Pusztai. This work is supported by the Olle Engkvist Foundation, the Knut and Alice Wallenberg Foundation and the European Research Council (ERC-2014-CoG grant 647121). Simulations were performed on resources at Chalmers Centre for Computational Science and Engineering (C3SE) provided by the Swedish National Infrastructure for Computing (SNIC).

Author contributions statement

Additional information

Competing interests: The authors declare no competing interests.