

Real-space Calculation of Orbital Hall Responses in Disordered Materials

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We developed an efficient numerical approach to compute the different components of the orbital Hall responses in disordered materials from the Berry phase theory of magnetization. We propose a theoretical framework based on the Chebyshev expansion of Green's functions and the position operator for systems under arbitrary boundary conditions. The capability of this scheme is illustrated by computing the orbital Hall conductivity for gapped graphene and Haldane model in presence of nonperturbative disorder effects. This methodology opens the door to realistic simulations of orbital Hall responses in arbitrary complex models of disordered materials.

Similarly to spintronics, *orbitronics* operate on stimuli capable of generating nonequilibrium distributions of orbital angular momentum^{1,2}. Seminal works from Bernevig *et al.* and Kontani *et al.* predicted the existence of an orbital Hall effect (OHE) in doped semiconductors³ and transition metals⁴, where a longitudinal current triggers a transverse flow of orbital angular momentum (OAM) independently of the spin-orbit coupling (SOC) of the material. Initially, the potential of the orbital degrees of freedom for technological applications has been questioned under the assumption of the quenching of the orbital angular momentum due to the crystal field⁵. However, recently, Go *et al.* demonstrated that even in systems with quenched orbital character, the application of electric fields enables hybridization channels absent at equilibrium, giving rise to the OHE⁶.

On the other hand, the experimental detection of the OHE was first investigated indirectly through orbital torque measurements^{7–12}, orbital pumping^{13,14}, and inverse orbital Hall effect^{15,16}. In these studies, the confirmation of the OHE remains debated due to the similarities in the symmetries obeyed by the OHE and the spin Hall effect, but two independent experimental groups have confirmed directly the existence of the OHE through magneto-optical Kerr rotation measurements in Ti¹⁷ and Cr¹⁸.

To date, most of those experimental results on the electrical generation of orbital currents and their application concentrate on three-dimensional (3D) systems. Nonetheless, the tunability in the properties of two-dimensional (2D) materials and the prospect of developing ultra-compact light-metal-based orbitronic devices has gained significant attention. For instance, theoretical works predicted that 1H transition metal dichalcogenides (TMDs) could host orbital-Hall insulating phases^{19,20} characterized by an orbital Chern number and in-gap OAM-carrying edge states²¹. Moreover, sizable OHE was predicted for various semiconducting 1T TMDs²². Additionally, the role of the orbital degrees of freedom in the generation of real-space localized non-equilibrium spin densities and their suitability for SOT was recently

revealed²³; and beyond TMDs, phosphorene was also identified as a suitable platform for unambiguous detection of orbital signals²⁴.

Aside from the muffin-tin picture that considers the contributions to the OAM from the immediacies of the atomic environment encoded in the orbital character of the wavefunctions, nonlocal contributions to the orbital moment arising from the electron wavepacket self-rotation addressed through the Berry phase description of the orbital magnetization^{25–29} can be dominant in systems with quenched orbital character such as gapped³⁰, bilayer³¹ and twisted bilayer graphene^{32,33}, and kagome lattices³⁴. Furthermore, Kazantsev *et al.* demonstrated that the net valley accumulation in gapped graphene nanoribbons and their associated orbital moments does not depend on the carrier diffusion lengths but on the characteristics of the electronic wave function near the edges³⁵. However, despite all the theoretical and experimental development, practical applications of orbitronics require an understanding of the role of the disorder in the generation of orbital currents and the relaxation of nonequilibrium orbital densities.

Examples of this necessity are the results from the experiments of Seifert *et al.* in magnetic bilayers, where magnetic disorder suppresses nonequilibrium spin densities and magnonic contributions, leaving their orbital currents and densities unaffected, which help in identifying a ballistic conduction of OAM¹⁵. On the other hand, Choi *et al.* reported an orbital diffusion length l_O , in Ti between 61–74 nm¹⁷ that contrasts with the short orbital diffusion length of $l_O = 6.6$ nm estimated by Lyalin *et al.* in Cr¹⁸. Such variation of l_O might be related to crystal field or disorder effects which remain mostly unexplored. The work of Pezo *et al.* suggests that random impurities decrease the value of the orbital Hall conductivity in Dirac materials³⁶, while other study reports that weak disorder can be the dominant source of current-induced orbital currents in the valence and conduction bands of Dirac materials³⁷. Conversely, for triangular lattices, the disorder appears to be detrimental³⁸.

In that context, usual diagonalization methods to treat

disorder effects become quickly prohibitive due to the computational cost scaling cubically with the system size. Accordingly, to further advance the field of orbitronics, there is a need for realistic, accurate, and efficient simulation tools capable of addressing non-perturbatively the role of the disorder in the generation of orbital currents in systems approaching experimentally relevant scales and geometries.

In this letter, we present a new efficient linear-scaling method that allows the computation of the orbital Hall conductivity (OHC) in disordered materials from the Berry phase theory of magnetization. Using the kernel polynomial method (KPM), we expand the position operator, the OAM, and use the Kubo-Bastin formula in terms of Chebyshev polynomials. We illustrate the methodology by studying the transport properties of orbital angular momentum in the topologically trivial and non-trivial phases of the Haldane model and investigate the effects of Anderson disorder. Our results suggest that including disorder favors the appearance of extrinsic Fermi -sea and -surface contributions that overcome the intrinsic orbital Hall responses for the clean case. The generality of the methodology enables the study of various current-induced orbital responses in disordered systems in a straightforward manner, independently of a basis choice for any temperature and chemical potential.

To address the components of the conductivity tensor, we use the linear response Kubo formalism. In the static limit, the elements of the OHC tensor are given by the Kubo-Bastin formula^{20,39,40}

$$\sigma_{\alpha\beta}^{\eta} = -\frac{i2\hbar e}{\Omega} \int_{-\infty}^{\infty} d\varepsilon F(\varepsilon) \text{ImTr} \langle J_{\alpha}^{\eta} \partial_{\varepsilon} G^{-}(\varepsilon, H) v_{\beta} \delta(\varepsilon - H) \rangle, \quad (1)$$

where Ω is the volume of the sample, v_{β} is the β component of the velocity operator $v_{\beta} \equiv \frac{i}{\hbar} [H, r_{\beta}]$, $G^{\pm}(\varepsilon, H) = \frac{1}{\varepsilon - H \mp i0\mp}$ is the retarded (advanced) Green's function, $F(\varepsilon)$ is the Fermi-Dirac distribution for a given temperature T and chemical potential μ , $J_{\alpha}^{\eta} = \frac{1}{2} \{L_{\eta}, v_{\alpha}\}$ is the orbital current operator. Though the eigenstate-based representation of the Kubo formula has been extensively used in the study of orbital angular momentum transport in clean systems^{1,21,30}, equation (1) has been instrumental in the investigation of electrical responses in disordered materials⁴¹⁻⁴³. Following the approach proposed by Bhowal and Vignale³⁰, we write the symmetrized orbital angular momentum operator as $\vec{L} = \frac{e\hbar}{4g_L\mu_B} (\vec{r} \times \vec{v} - \vec{v} \times \vec{r})$, where g_L is the Landé g -factor for the orbital angular momentum, and μ_B is the Bohr magneton. From this definition, it is clear that the difficulties in computing the orbital angular momentum beyond the muffin-tin approximation are related to the ill-defined nature of the position operator in periodic systems. Nonetheless, as noted by Bianco and Resta⁴⁴, even if the diagonal elements in the energy eigenstates

of the position operator are ill-defined, the off-diagonal elements can be easily written as $\langle i | r_{\alpha} | j \rangle = i\hbar \frac{\langle i | v_{\alpha} | j \rangle}{E_j - E_i}$. Thus, using the definition of Green's functions, we define equivalent representations of the same position operator as

$$\begin{aligned} \langle i | r_{\alpha}^{\text{I}} | j \rangle &= i\hbar \langle i | \int d\varepsilon' [G^{+}(\varepsilon', H) v_{\alpha} \delta(H - \varepsilon')] | j \rangle \\ \langle i | r_{\alpha}^{\text{II}} | j \rangle &= -i\hbar \langle i | \int d\varepsilon' [\delta(H - \varepsilon') v_{\alpha} G^{-}(\varepsilon', H)] | j \rangle \end{aligned} \quad (2)$$

These representations of the r_{α} operator in (2) are formally equivalent in the limit of vanishing broadening. However, for their numerical evaluation, both versions of the operator must be considered to cancel any spurious contributions due to the regularization of Green's functions. The numerical evaluation of equation (2) is inefficient if one uses the eigenstates of the system, at contrast with spectral methods such as the KPM which perform computation as matrix-vector operations. Following the Chebyshev expansion technique^{42,45}, we rescale the Hamiltonian H and the energies ε between the $(-1, 1)$ interval and expand the Green's and spectral functions as a polynomial series of the rescaled Hamiltonian $\tilde{H}$ and energies $\tilde{\varepsilon}$. Thus the expanded expression for r^{I} in (2) is

$$\begin{aligned} \langle i | r_{\alpha}^{\text{I}} | j \rangle &= i\hbar \frac{2}{\Delta E} \int_{-1}^1 d\tilde{\varepsilon}' \langle i | \left(\sum_{\mu} c_{\mu}(\tilde{\varepsilon}') T_{\mu}(H) \right) v_{\alpha} \\ &\quad \times \left(\sum_{\nu} f_{\nu}(\tilde{\varepsilon}') T_{\nu}(\tilde{H}) \right) | j \rangle. \end{aligned} \quad (3)$$

The coefficients $c_{\mu}(\varepsilon) = \frac{-2i}{\sqrt{1-\varepsilon^2}} \frac{g_{\mu} e^{i\mu \arccos(\varepsilon)}}{(\delta_{\mu,0}+1)}$ and $f_{\nu}(\varepsilon) = \frac{2}{\pi\sqrt{1-\varepsilon^2}} \frac{g_{\nu} T_{\nu}(\varepsilon)}{(\delta_{\nu,0}+1)}$ are related to the polynomial expansion of $G^{+}(\varepsilon, H)$ and $\delta(H - \varepsilon)$, respectively⁴⁶, ΔE is the energy width of the spectrum defined as $\Delta E = (E_{\text{max}} - E_{\text{min}})/2$, $T_m(x) = \cos(m \arccos(x))$ is the m -th Chebyshev polynomial of first-kind and g_{μ} is a damping factor added to the series to smooth the Gibbs oscillations resulting from the truncation of the polynomial expansion at order M ⁴⁵. The components of r^{II} are obtained from the relation $r_{\alpha}^{\text{II}} = (r_{\alpha}^{\text{I}})^{\dagger}$. Using these expansions, the orbital angular momentum operator L_{η} is written as $L_{\eta} = \epsilon_{\alpha\beta\eta} \frac{e\hbar^2}{4g_L\mu_B} (r_{\alpha}^{\text{I}} v_{\beta} - v_{\alpha} r_{\beta}^{\text{II}}) = \frac{e\hbar^2}{4g_L\mu_B} \ell_{\eta}$, with $\epsilon_{\alpha\beta\eta}$, being the Levi-Civita symbol. Inserting the expansion of the L_{η} operator into (1) and using the polynomial expansion of the Green and spectral functions, the components of the OHC tensor reads as:

$$\begin{aligned} \sigma_{\alpha\beta}^{\eta} &= -\frac{8}{\Delta E^3} \frac{ie^2\hbar^3}{4g_L\mu_B\Omega} \\ &\quad \times \int_{-1}^1 d\tilde{E} \frac{F(\tilde{E})}{(1-\tilde{E})^2} \sum_{m,n} 2\mu_{m,n}^{\alpha,\beta,\eta} \text{Im} \left(\Gamma_{m,n}(\tilde{E}) \right), \end{aligned} \quad (4)$$

$\mu_{m,n}^{\alpha,\beta,\eta} = \frac{g_m g_n}{(1+\delta_{m,0})(1+\delta_{n,0})} \text{Tr} \langle \frac{1}{2} \{ \ell_\eta, v_\alpha \} T_m(\tilde{H}) v_\beta T_n(\tilde{H}) \rangle$ contains all the information related to the Hamiltonian and the orbital current operator. It neither depends on the energy nor the specific basis chosen for describing the system and comprises the most expensive part of the calculation (details on the computation of these coefficients are shown in SM⁴⁷). In contrast, $\Gamma_{m,n}(\tilde{E})$ is an energy-dependent scalar that does not depend on the specific features of the Hamiltonian and reads as

$$\Gamma_{m,n}(\tilde{E}) = \left(\tilde{E} + im\sqrt{1-\tilde{E}^2} \right) e^{-im \arccos(\tilde{E})} T_n(\tilde{E}). \quad (5)$$

Equation (4) constitutes the main result of our work. After determining the coefficient matrix $\mu_{m,n}^{\alpha,\beta,\eta}$, we can compute the energy-resolved OHC for all the temperatures. To evaluate the traces, we use the random phase approximation which allows the expansion of the trace in terms of a subset of elements⁴⁵.

To illustrate the method, we apply it to study the transport of orbital currents in the Haldane model⁴⁸. The Hamiltonian is given as $H = -t \sum_{\langle i,j \rangle} c_i^\dagger c_j + i \frac{t_2}{3\sqrt{3}} \sum_{\langle\langle i,j \rangle\rangle} \nu_{ij} c_i^\dagger c_j + \sum_i (\Delta_i + \xi_i) c_i^\dagger c_i$, where t and t_2 are the first and second nearest neighbor hoppings, respectively. $\nu_{ij} = +1(-1)$ if the second nearest neighbor hopping from the sites j to i occurs in the counter-clockwise (clockwise) direction. $\Delta_i = \Delta(-\Delta)$, is the onsite energy for $i \in A(i \in B)$ sublattice and ξ_i is an on-site Anderson disorder term whose values are selected from a uniform random distribution in the interval $[-W/2, W/2]$. Following Ref. 30, we fix $t = 2.8$ eV and the carbon-carbon distance $a = 1.42$ Å.

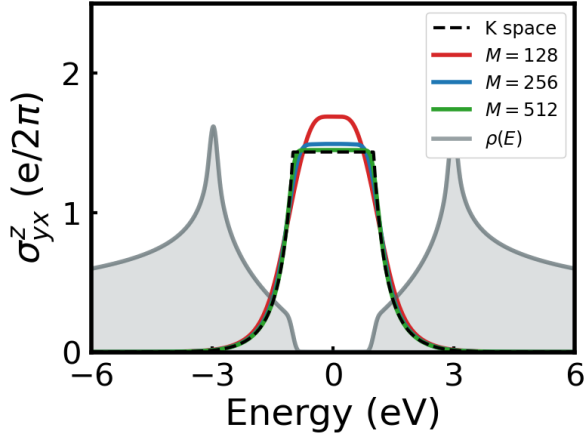


FIG. 1. Real-space orbital Hall conductivity of the Haldane model with $t_2 = 0$, $\Delta = 1.0$ eV, and $W = 0$ (for different number of moments M , and compared with the diagonalization result (dashed line)). The shaded curve represents the DOS computed for $M=512$.

Our starting point is the study of the convergence of the OHC plateau for various values of M . Figure 1 shows the orbital Hall conductivities obtained using the reciprocal space procedure described in Ref. 30 and our real-space method for clean gapped graphene ($t_2 = 0$). We

consider systems composed of 256×256 unit cells with an onsite potential $\Delta = 1$ eV, using the Jackson kernel. As the figure shows, for smaller values of M the orbital-Hall conductivity plateau is broadened and it goes to a slightly higher value than the exact (reciprocal-space computed) OHC plateau. Nonetheless, as M increases the agreement between the two methods increases remarkably. The improvement in the description is due to the reduction of the numerical broadening associated with the Chebyshev polynomial expansion, which for the Jackson kernel decreases as $\sim 1/M$ ^{42,45}.

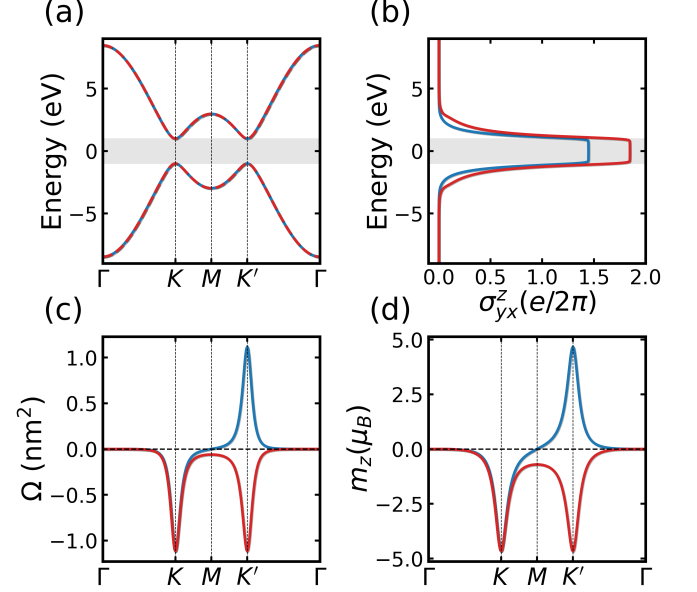


FIG. 2. (a) Energy bands of the Haldane model in the trivial (blue lines) phase with $\Delta = 1.0$ eV and $t_2 = 0$, and the nontrivial (red lines) phase $t_2 = 1.0$ eV and $\Delta = 0$. (b) Orbital Hall conductivity for the Haldane model in the trivial and nontrivial phases, computed for systems with 256×256 unit cells and $M = 512$. (c) Berry curvature of the lowest energy band of the Haldane model in the trivial and nontrivial topological phases. (d) Orbital magnetic moment for the lowest energy band of the Haldane model in both phases.

Following this, we study the interplay between nontrivial topologies and the transport of orbital currents. To make a fair comparison, we set the energy gaps of the trivial and nontrivial phases to have the same value (see FIG. 2 (a)) and compute their OHC. FIG. 2 (b) shows the results of the calculations. Upon inspection, it is clear that the OHC for both cases does not exhibit quantized values due to the nonconserved nature of the orbital angular momentum, with the topologically nontrivial case having a slightly larger OHC than the trivial phase. As Refs. 30, 34, 49, and 50 pointed out, the transport of the orbital angular momentum in systems with quenched orbital character occurs solely due to the combined action of the Berry curvature and the orbital moment distribution in reciprocal space. Thus, it is clear that the simultaneous inversion of the Berry curvature and orbital

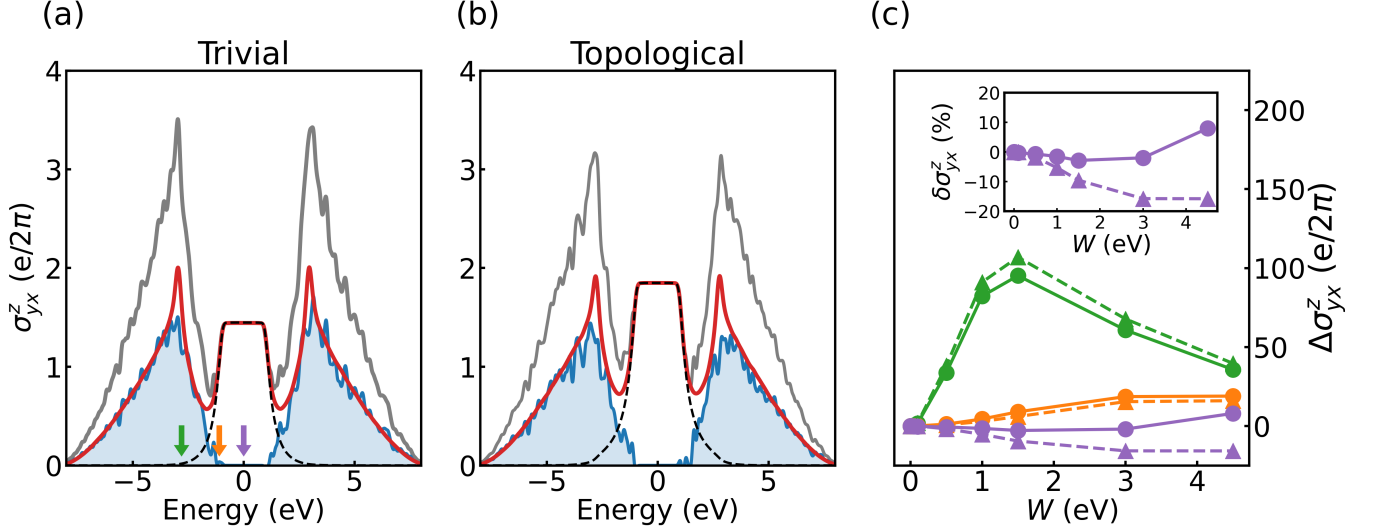


FIG. 3. Total (grey line), Fermi sea (red line) and Fermi surface (blue shaded area) contributions of the orbital Hall conductivity for the Haldane model in the trivial phase for $t_2 = 0$, $\Delta = 1.0$ eV, and $W = 0.1$ eV (a), and the topologically nontrivial phase for $t_2 = 1.0$ eV, $\Delta = 0.0$, and $W = 0.1$ eV (b) computed for systems with 256×256 unit cells, $M = 512$ and averaged over 72 disorder configurations. The case for $W = 0.0$ (dashed line) is shown as a guide to the eye. (c) Variation of the orbital Hall conductivity with W for the three energies highlighted in panel (a), the solid and dashed lines correspond to the trivial and topologically nontrivial phases of the Haldane model, respectively. Inset: Percentual variation of the orbital Hall conductivity plateau with respect to the case with $W = 0$ for the topologically trivial and nontrivial phases.

moment that occurs in the trivial phase due to the time-reversal symmetry (see FIG. 2 (c) and FIG. 2 (d)) leads to an overall smaller OHC for the topologically trivial case. These results highlight the dependence of the orbital Hall conductivity on the geometric features of the energy states of the system through the Brillouin zone and add up to the previous theoretical evidence showing that in topologically nontrivial phases, such as the quantum spin-Hall and quantum anomalous-Hall phases, the topologically protected edge states can carry finite orbital angular momentum³⁹.

We further investigate the effects of Anderson disorder on the OHC in both trivial and topologically nontrivial phases of the Haldane model. To highlight the modifications to the OHC induced by the disorder, we use the decomposition of the Kubo formula proposed by Bonbien and Manchon⁵¹ to separate the Fermi sea and Fermi surface contributions to the total OHC. FIG. 3 (a) and FIG. 3 (b) show the OHC for the trivial and topological phases, respectively, for $W = 0.1$ eV (curves for other values of W are shown in the supplementary material⁴⁷), together with the clean cases as a guide to the eye (dashed lines). From the two figures, it is clear that the orbital-Hall conductivity plateaux remain unchanged by the disorder, indicating its robustness. Aside from this, outside the energy gap, peaks in the OHC in both the Fermi surface and sea contributions appear and attain their maximum value at energies comparable to t . To inquire about these disorder-enabled effects, we tracked the evolution of the OHC as a function of W for three energies of interest signaled by the arrows in

FIG. 3 (a), which correspond to the middle of the energy gap (purple arrow), 100 meV below the lowest gap edge (orange arrow) and -2.8 eV (green arrow). FIG. 3 (c) shows the changes in the Fermi sea contributions to the OHC at these energies, where the solid and dashed lines correspond to the trivial and the topologically nontrivial phases, respectively. As seen in the figure, contributions at $E = -2.8$ eV increase monotonically with W , and saturate for $W \sim 1.5$ eV, where multiple scattering and localization effects start to dominate and reduce the OHC. In contrast, the OHC plateau presents small changes with the disorder. The inset illustrates this better by showing the percentual change concerning the clean case. Here, it shows that for W between $[0, 1]$ eV, the change in the height of the plateau is below 5% with the plateau for the topological case decreasing in contrast with the trivial case. Increasing values of W accentuate this tendency, and the OHC for the topological case is reduced by almost 20% of its height, and for the trivial case, it is increased by 10%. The permanence and relatively small changes in the OHC go along with the analysis of Bernevig *et al.*³. Moreover, our results show some discrepancies with the tendencies presented by Pezo *et al.*; we attribute this to the small size of their systems³⁶. Regarding the increasing contributions at $E = -2.8$ eV, we argue that they might be related to the formation of disorder-enabled current loops similar to those previously observed in graphene nanoribbons⁵² and quantum dots⁵³, which have been overlooked in this type of calculations due to the difficulties in addressing the position operator in systems with periodic boundaries.

In conclusion, we have presented a numerical method based on the Chebyshev expansion technique to evaluate the components of the orbital Hall conductivity tensors as described by the Berry phase formulation of the orbital magnetization. Combining Green's function theory with the KPM, we obtained a systematic form to evaluate the position operator whatever the boundary conditions. We have validated and illustrated the capability of the method through the analysis of the Haldane model and disorder effects on the transport of orbital angular momentum in its topologically trivial and nontrivial phases. Our findings evidence that the orbital Hall conductivity plateau is robust and demonstrate that for graphene-like systems, the effects of disorder break the symmetries of the systems while creating new channels for the electrical generation of orbital currents within the bulk of the systems, in analogy with prior results in graphene nanoribbons. This novel approach enables the real-space investigation of the role of the disorder in the orbital angular momentum transport and other position-dependent observables in disordered systems of arbitrary complexity.

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