Supplementary Material for

Chirality Induced Spin Selectivity: A Classical Spin-off

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This supplementary information document includes the following sections:

Section 1. Initial position sensitivity test

To produce the results presented in the main text we initially positioned the center of mass (COM) of the charged C_{60} molecules at the top of the helical channel, 8 Å above the lowest point of the circular cross-section, along the vertical (z) direction. To verify that the qualitative nature of our results and the corresponding conclusions do not strongly depend on these initial conditions, we repeated some of the simulations with initial heights of 12, 30, and 45 Å (below, at, and above the center of the circular cross-section, respectively, see SI movie 2). Fig. S1 exhibits the molecular trajectories as a function of time for the three additional initial positions considered with a kinetic friction coefficient of μ_k = 0.1 under a static vertical electric field intensity of $E_z = 0.05$ V/Å. All trajectories exhibit the same qualitative trends with some quantitative differences for the clockwise rotating molecules (see Fig. S1a). The spatial separation is enhanced with increasing initial height due to the higher impact velocity of the molecule with the frictional wall (see Fig. S1b).

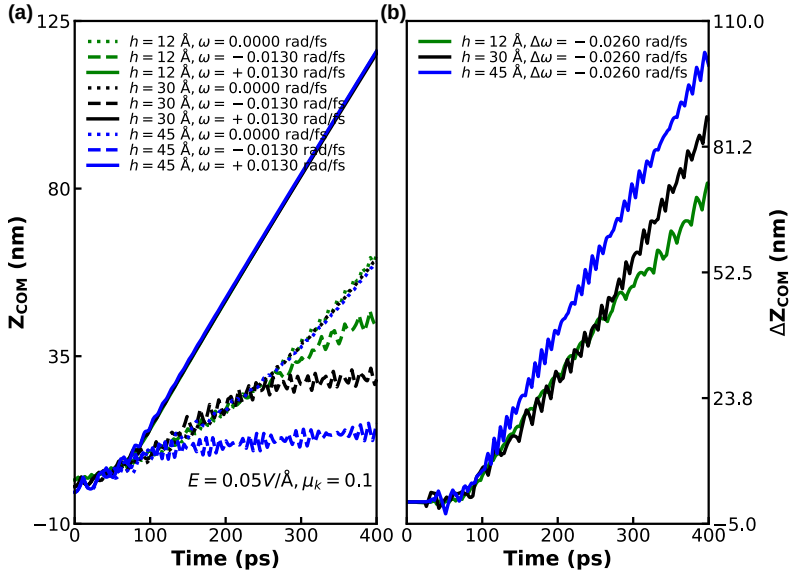

FIG. S1. Effect of the release height on the trajectory of initially spinning C_{60} molecules traversing a right-handed helical tube. (a) Time evolution of the vertical COM position of counterclockwise ($\omega_z = +0.013$ rad/fs, solid lines) and clockwise ($\omega_z = -0.013$ rad/fs, dashed lines) spinning charged ($q = -1$ a.u.) C_{60} molecules released at a height of h = 12 (green lines), 30 (black lines), and 45 (blue lines) Å, accelerated by a vertical electric field of 0.05 V/Å and experiencing a kinetic friction coefficient of $\mu_k = 0.1$ by the tube wall. Results of non-rotating molecules ($\omega_z = 0$ rad/fs) are presented for comparison by the dotted lines. (b) Time evolution of the vertical COM separation between C_{60} molecules initially spinning in opposite senses while being released at different initial heights. Line colors match those presented in panel (a).

We further examined the effect of lateral shifts of the initial C_{60} COM position on the molecular trajectories. Fig. S2 presents simulation results for an initial COM height of 30 Å laterally shifted within the helical tube by $\Delta x = \pm 10$ and ± 20 Å. Here, as well, all trajectories exhibit the same qualitative trends with some quantitative differences for the clockwise rotating molecules (see Fig. S2a). Specifically, positive lateral shifts are found to produce larger long-term molecular deceleration, whereas negative lateral shifts produce lower deceleration. This may be attributed to the fact that positive shifts cause the molecule to encounter the surface at a lower curvature region thus enhancing its deceleration when spinning in the clockwise sense, whereas negative lateral shifts drive the molecule towards a higher curvature surface thus reducing its deceleration. An opposite effect occurs for the counterclockwise spinning molecules but is less pronounced due to their higher COM velocity which reduces the collision time. Altogether, the spatial separation dynamics between oppositely spinning molecules exhibits minor dependence on the initial lateral position of the molecule with mild enhancement (reduction) of the effect for positive (negative) shifts (see Fig. S2b).

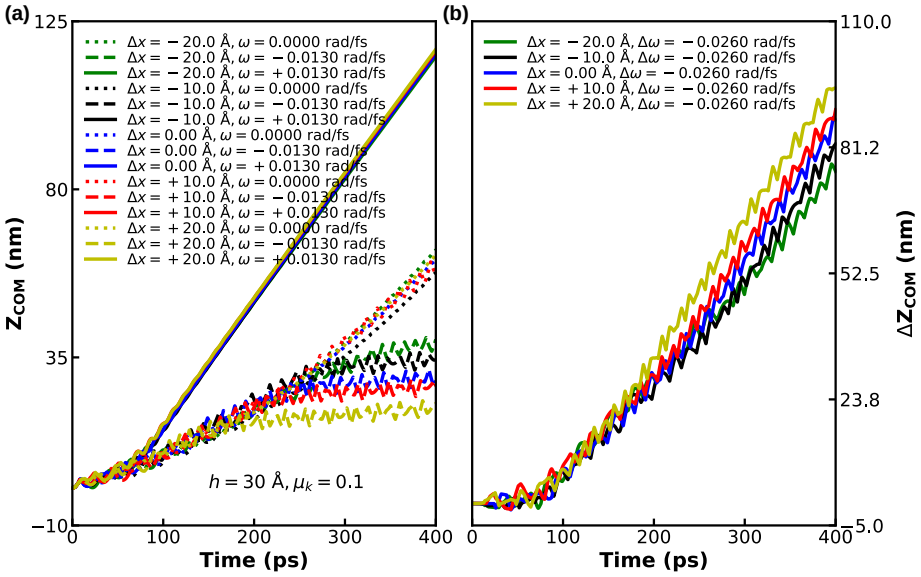

FIG. S2. Effect of lateral shift on the trajectory of initially spinning C_{60} molecules traversing a right-handed helical tube. (a) Time evolution of the vertical COM position of counterclockwise ($\omega_z = +0.013\ rad/fs$, solid lines) and clockwise ($\omega_z = -0.013\ rad/fs$, dashed lines) spinning charged ($q = -1\ a.u.$) C_{60} molecules released at an initial height of $h =$ 30 Å with lateral shifts of $\Delta x = 0$ (blue lines), ± 10 (red/black lines), ± 20 (yellow/green lines) Å, accelerated by a vertical electric field of $0.05\ V/$Å and experiencing a kinetic friction coefficient of $\mu_k = 0.1$ by the tube wall. Results of non-rotating molecules ($\omega_z = 0\ rad/fs$) are presented for comparison by the dotted lines. (b) Time evolution of the vertical COM separation between C_{60} molecules initially spinning in opposite senses with different initial lateral shifts. Line colors match those presented in panel (a).

Section 2. Time-step convergence tests

To ensure that our results are converged with respect to the choice of fixed molecular dynamics simulation time-step, we repeated some of the simulations using time steps that are an order of magnitude larger or lower than the original value of $\Delta t = 0.001$ fs used to obtain the results presented in the main text, while keeping all other parameters the same. Fig. S3 compares the COM C_{60} trajectories with friction coefficients of $\mu_k = 0$ (a), 0.1 (b), 0.4 (c), and 0.8 (d) calculated with a time step of $\Delta t = 0.01$ (black), 0.001(blue), and 0.0001 (red) fs. For kinetic friction coefficients lower than 0.4, negligible differences between the trajectories obtained using the different time-steps are observed. At the highest friction coefficient considered of $\mu_k = 0.8$, following the initial dynamics, some long-term quantitative deviations are observed, especially for the larger time-step considered, with no impact on the qualitative nature of the predictions. Figs. S4 and S5 present similar analyses for the time-step sensitivity of the angular velocity dependence and driving electric field intensity dependence of the molecular trajectories, further demonstrating that our choice of simulation time-step provides well-converged results.

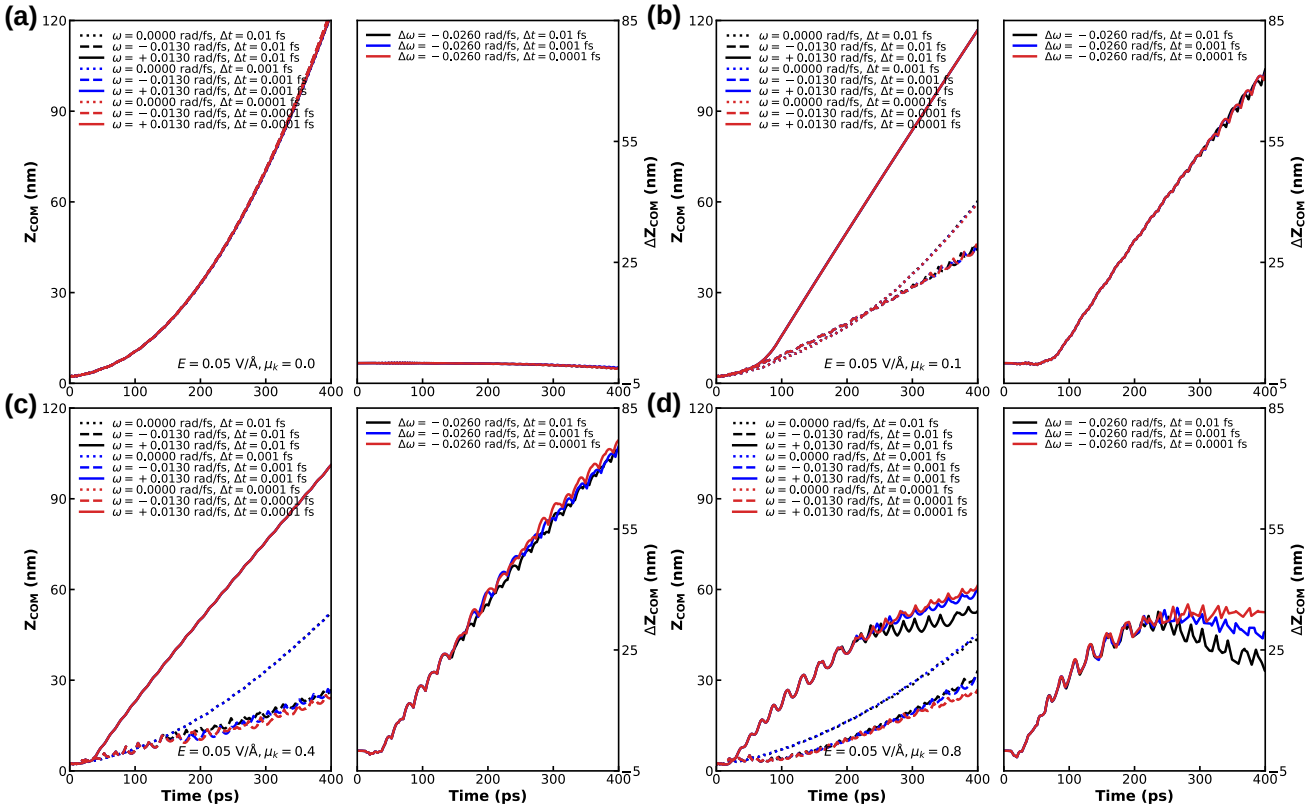

FIG. S3. Sensitivity tests of the friction-coefficient dependence of the molecular trajectories with respect to the simulation time-step. Left panels: Time evolution of the vertical COM position of counterclockwise ($\omega_z = +0.013\ rad/fs$, solid lines) and clockwise ($\omega_z = -0.013\ rad/fs$, dashed lines) spinning charged ($q = -1\ a.u.$) C_{60} molecules, accelerated by a vertical electric field of $E_z = 0.05\,V/\text{Å}$ and experiencing kinetic friction coefficients of $\mu_k = 0.0$ (a), 0.1 (b), 0.4 (c), and 0.8 (d) by the tube wall. Results of non-rotating molecules ($\omega_z = 0\ rad/fs$) are presented by the dotted lines. Right panels: Corresponding time evolution of the vertical COM separation between C_{60} molecules initially spinning in opposite senses. Results obtained with time-steps of $\Delta t = 0.01$, 0.001, and 0.0001 fs, are marked by black, blue, and red lines, respectively.

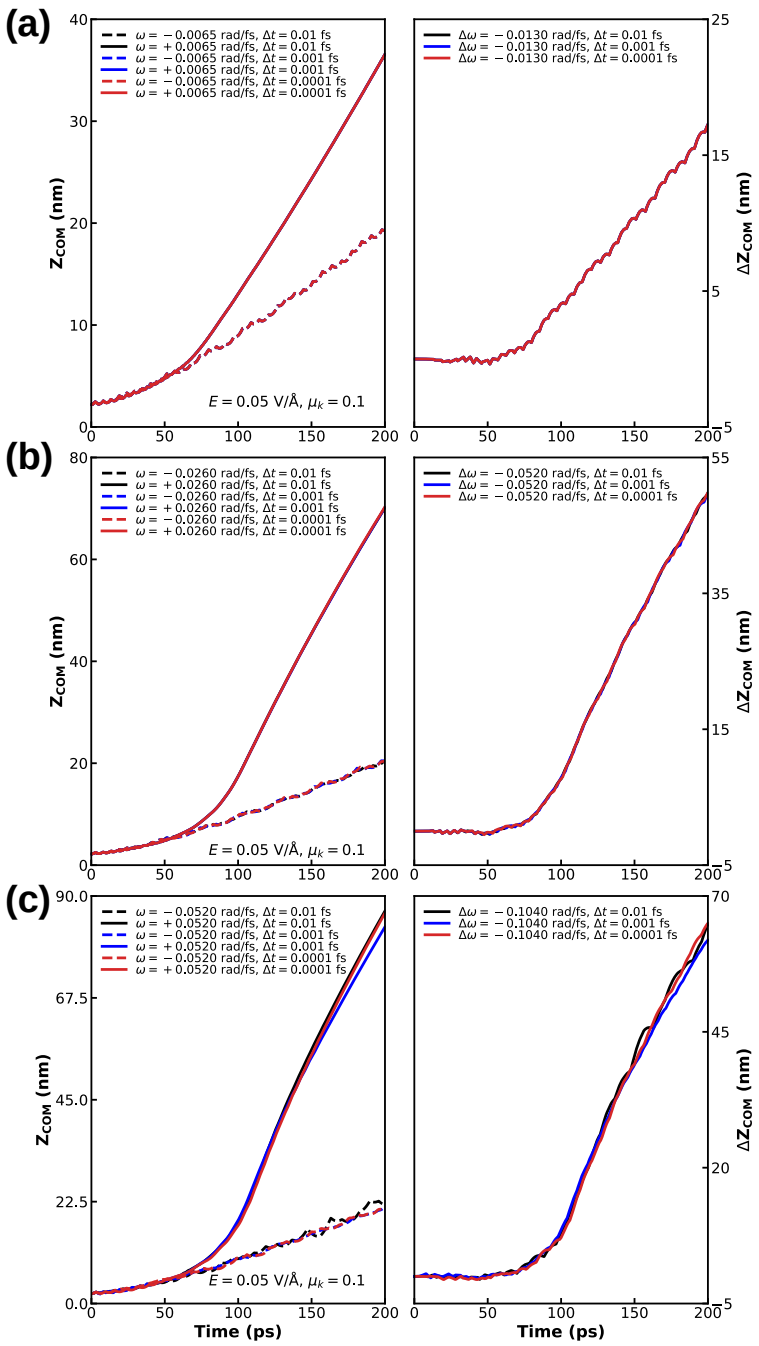

FIG. S4. Sensitivity tests of the angular velocity dependence of the molecular trajectories with respect to the simulation time-step. Left panels: Time evolution of the vertical COM position of counterclockwise (solid lines) and clockwise (dashed lines) spinning C_{60} molecules with initial angular velocities of $\omega_z = \pm 0.0065$ (a), ± 0.0260 (b), and ± 0.0520 (c) rad/fs. The charged ($q = -1\ a.u.$) molecules are accelerated by a vertical electric field of $E_z = 0.05\,V/$Å and experience a kinetic friction coefficient of $\mu_k = 0.1$ by the tube wall. Right panels: Corresponding time evolution of the vertical COM separation between C_{60} molecules initially spinning in opposite senses. Results obtained with time-steps of $\Delta t = 0.01$, 0.001, and 0.0001 fs, are marked by black, blue, and red lines, respectively.

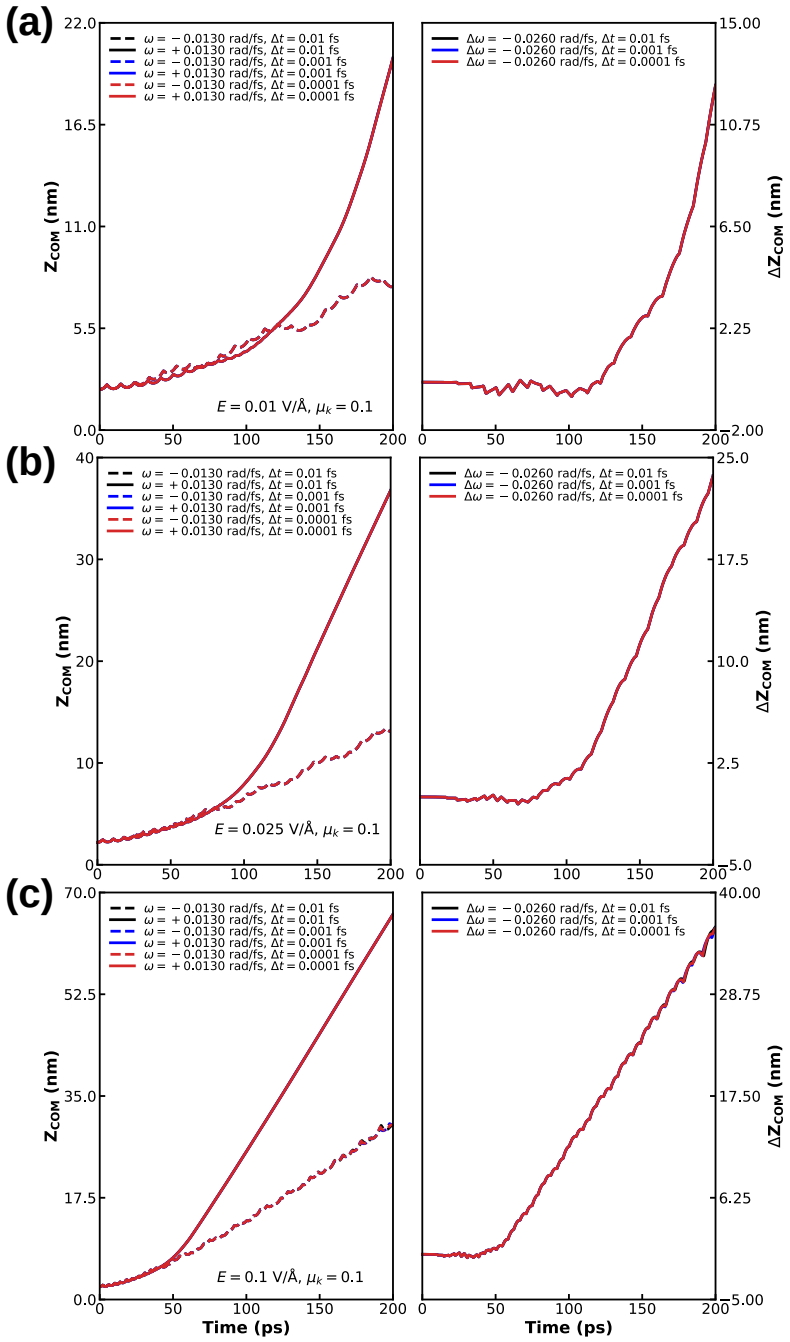

FIG. S5. Sensitivity tests of the driving electric field intensity dependence of the molecular trajectories with respect to the simulation time-step. Left panels: Time evolution of the vertical COM position of counterclockwise (solid lines) and clockwise (dashed lines) spinning C_{60} molecules with an initial angular velocity of $\omega_z = \pm 0.013$. The charged ($q = -1\ a.u.$) molecules are accelerated by vertical electric fields of $E_z = 0.01$ (a), 0.025 (b), and 0.1 (c) $V/\text{Å}$ and experience a kinetic friction coefficient of $\mu_k = 0.1$ by the tube wall. Right panels: Corresponding time evolution of the vertical COM separation between C_{60} molecules initially spinning in opposite senses. Results obtained with time steps of 0.01, 0.001, and 0.0001 fs, are marked by black, blue, and red lines, respectively.