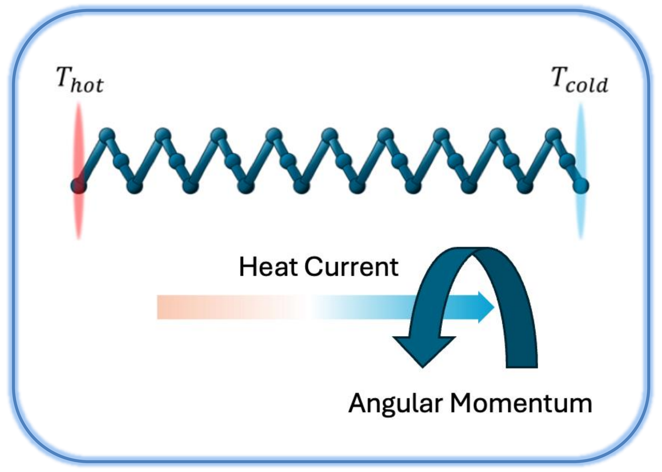
Consequences of Linear-Angular Momentum Coupling in Chiral Molecular Systems

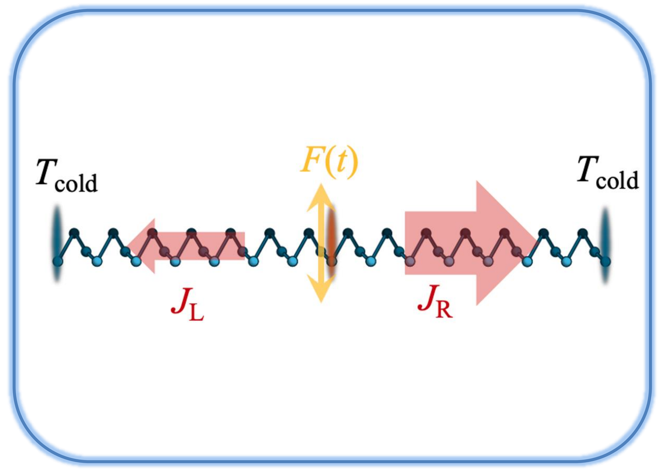
Jichen Feng, Ethan Abraham, Joseph E. Subotnik, Abraham Nitzan
University of Pennsylvania

Background: This poster covers two works

- The consequences of linear-angular momentum correlation (locking) in the behavior of electronic transport in a chiral environment have been under study for some time. More recently, the angular momentum of nuclear motions (so-called chiral phonons) has also come into focus.
- Out-of-equilibrium driving can bias the population of modes with a preferred handedness, enabling **angular-momentum generation** and **directional energy flow**.
- Two complementary preprints are summarized here: (1) **thermal-current-induced** angular momentum and (2) **phase-controlled** vibrational rectification.

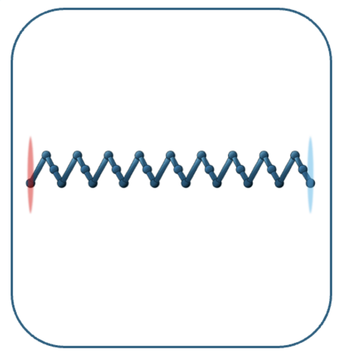


Thermal bias: chiral chain between hot/cold baths



Coherent drive: phase ϕ selects polarization and sets J/J .

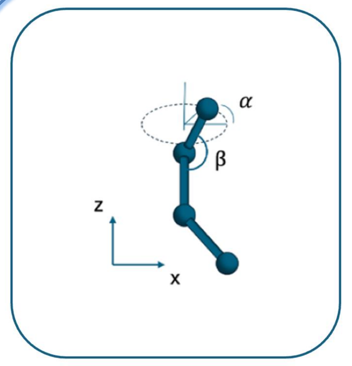
Models in our works



Helical Chain

$$H_A=\sum_{j=1}^{N}\frac{1}{2}m\dot{\vec{x}}_j^2+\sum_{j=1}^{N-1}\frac{1}{2}(\vec{x}_j-\vec{x}_{j+1})^T\mathbf{K}_{j,j+1}(\vec{x}_j-\vec{x}_{j+1})+\sum_{j=1}^{N}\frac{1}{2}m\omega_j^2\vec{x}_j^2$$

A harmonic helical chain, earlier used by Chen et al. [1] to study how helical structure manifests in the correlation between linear and angular momentum.

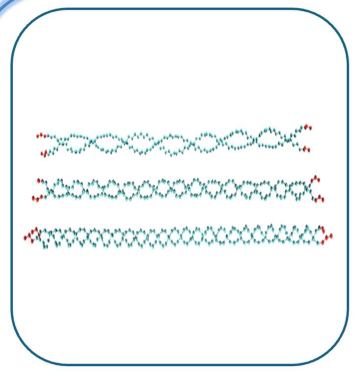


4-Atom Molecule

$$H_B=\sum_{j=1}^{4}\frac{1}{2}m\dot{\vec{x}}_j^2+V_B(\{\vec{x}_j\})$$

$$V_B(\{\vec{x}_j\})=\sum_{j=1}^{3}\frac{1}{2}K\left[|(\vec{x}_j+\vec{r}_j)-(\vec{x}_{j+1}+\vec{r}_{j+1})|-r_0\right]^2+\sum_{j=1}^{4}\frac{1}{2}m\omega_0^2\vec{x}_j^2$$

A 4-atom entity whose chirality is determined by how the bond and dihedral angles are set.



Polyethylene Double-Helix

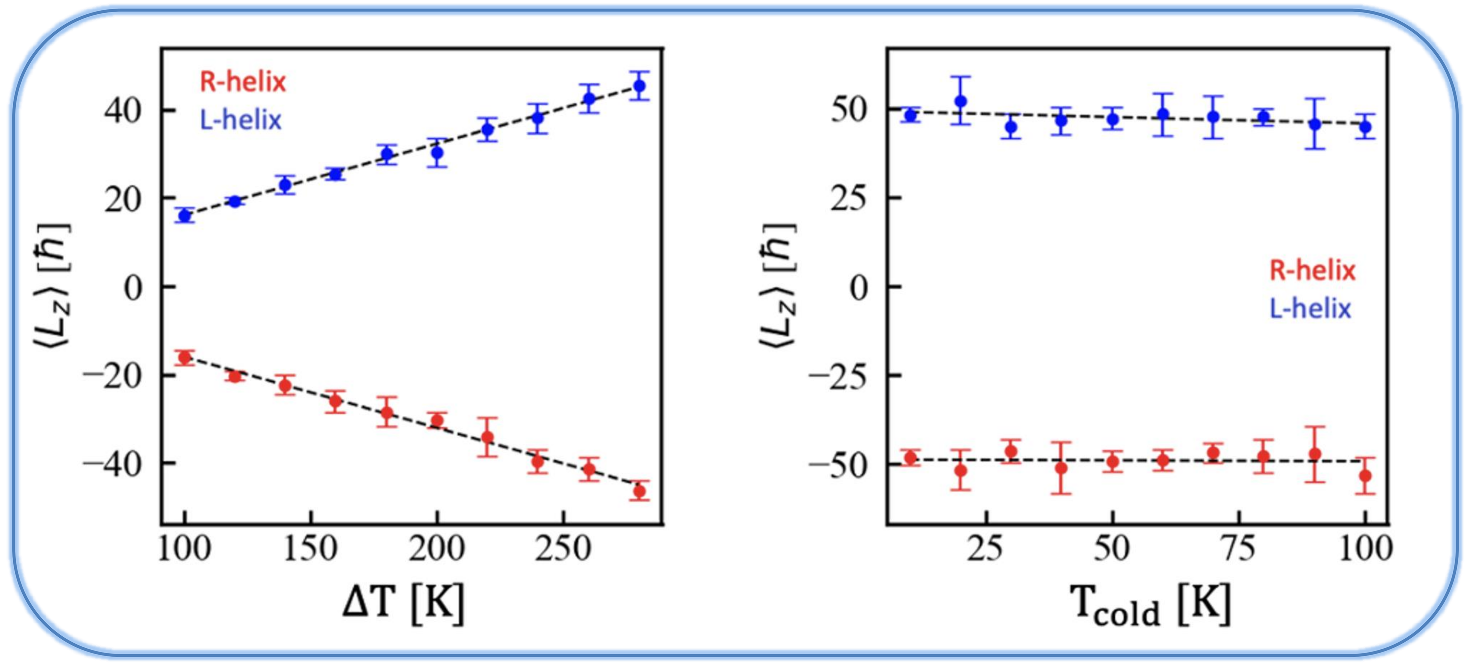
$$H_C=\sum_{j=1}^{2N}\frac{1}{2}m\dot{\vec{x}}_j^2+\sum_{j=1}^{2N-2}k_b(l_j-l_0)^2+\sum_{j=1}^{2N-4}k_\theta(\theta_j-\theta_0)^2+\sum_{j=1}^{2N-6}\sum_{n_j}^{4}\frac{C_n}{2}[1+(-1)^{n_j-1}\cos(n_j\phi_j)]+\sum_{j=1}^{2N}\sum_{i\neq j}4\varepsilon_{ij}\left[(\frac{\sigma}{r_{ij}})^{12}-(\frac{\sigma}{r_{ij}})^{6}\right],$$

A twisted double-helix structure, which is more realistic.

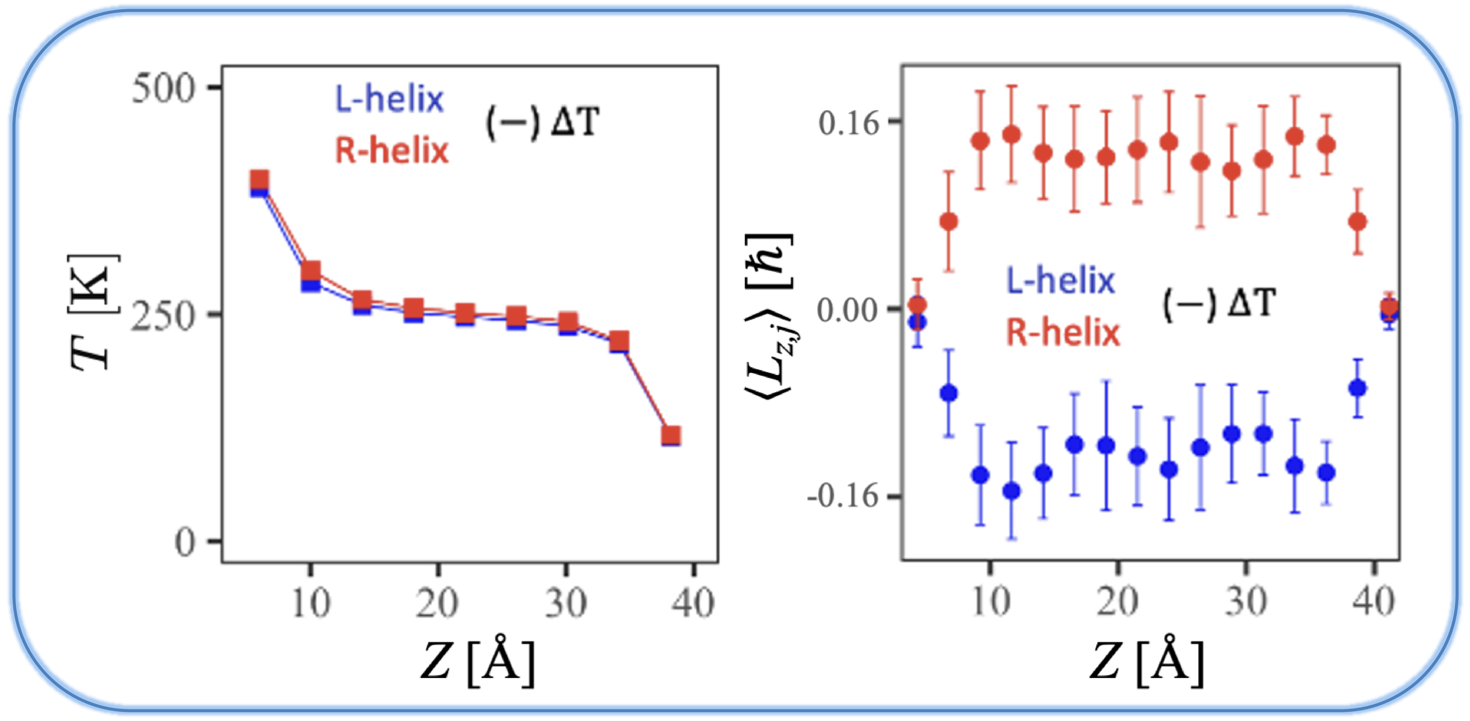
1.Chen, H.; Wu, W.; Zhu, J.; Yang, Z.; Gong, W.; Gao, W.; Yang, S. A.; Zhang, L., Chiral Phonon Diode Effect in Chiral Crystals. *Nano Letters* **February 11, 2022**.

Nuclear Angular Momentum Generation in Thermally Driven Chiral Systems

- We connected the models between hot and cold baths with different temperature configurations and studied the correlation between the angular momentum generations and temperature profiles.



- The results displayed above show numerical simulation results for the harmonic helical chain model with polymerization N = 201. The generated angular momentum strongly depends on the temperature difference ΔT(left) that induces the heat current J_H and only weakly on the average temperature (right).

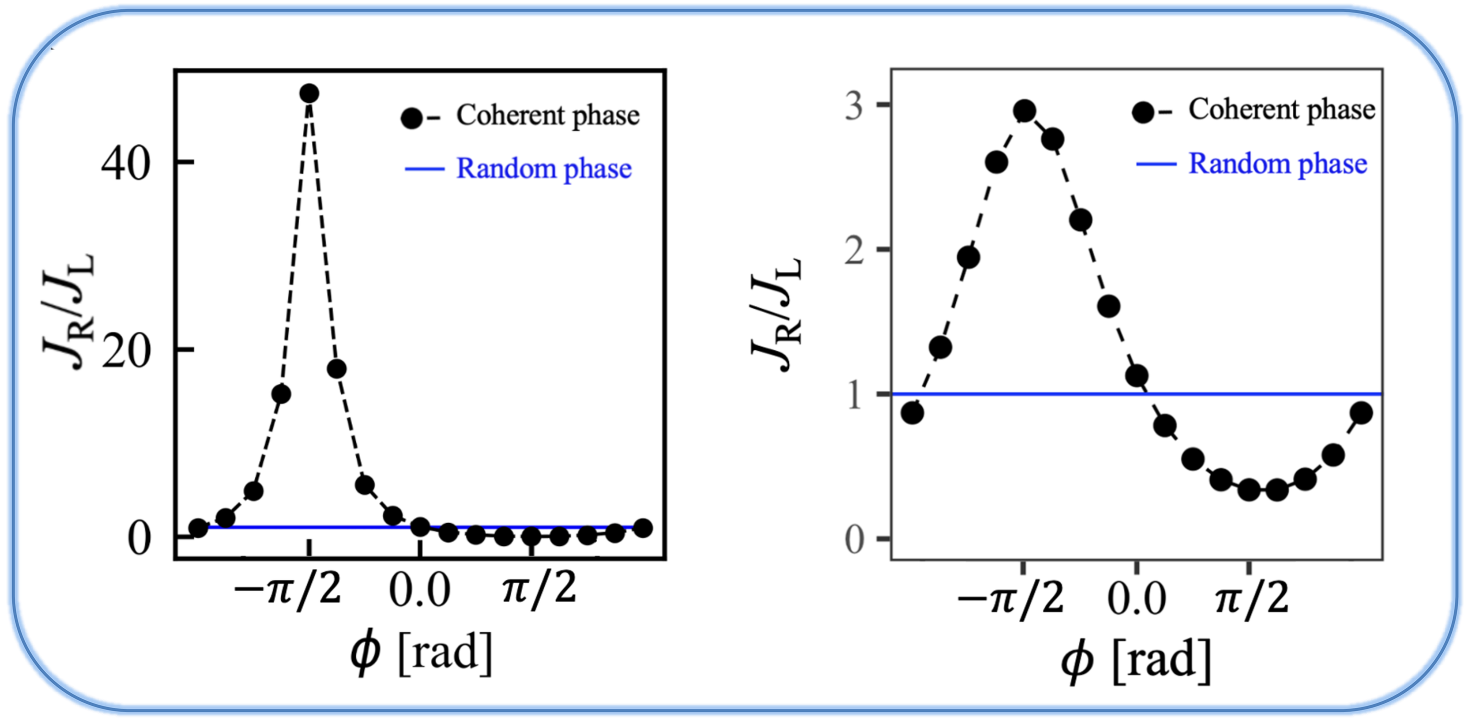


- The angular momentum generation phenomenon is robust and remains in the double helix model. Above shows the temperature and angular momentum profile in the system, with different handedness.
- Below is a table discussing the amplitude of the angular momentum generation, comparing our work with the generation from circularly polarized light (CPL).

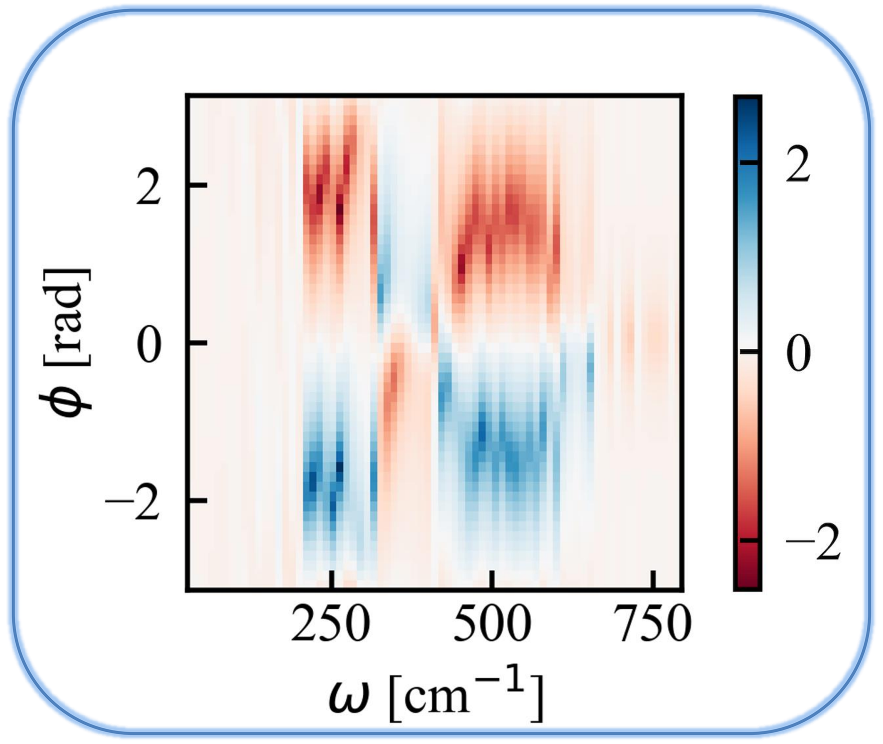
Feature	CPL Excitation	Thermal Excitation (This Work)
Angular momentum per atom	$\sim\hbar/10$	$\sim\hbar/10$ (simulated in this work)
Duration	< 1 ps; ultrafast transient	Steady-state
Coherence	Coherent (but decoheres quickly)	Incoherent
Experimental complexity	High: CPL source, tuning	Moderate: heat flux via contacts
Chirality origin	External—chiral perturbation	Internal—chiral material
Direction control	CPL handedness	Enantiomer or heat flux direction
Frequency resolution	Light frequency	Filtered thermal driving
System size scaling	Confined to optical penetration depth	Scales with system length $\propto L$

Rectification of Vibrational Energy Transfer in Driven Chiral Molecules

- In the simulation, we applied a coherent driven force at the center of the chain and explored the heat rectification behavior in the helical chain and double helix systems.



- The figure above displays numerical simulation results for the helical chain model (left) and double helix model (right). the greatest difference from unity when the driving phase is equal to $\pm\pi/2$, and that reversing the phase ($\phi\rightarrow-\phi$) results in the reciprocal rectification ratio ($J_R/J_L\rightarrow J_L/J_R$).



Left plots shows the heatmap of the rectification $Log_{10}(J_L/J_R)$ vs driving phase ϕ and frequency ω. The phase dependence of the rectification effect is related to the structure of chiral phonon modes and the frequency dependence results from both the polarization of the phonon band structure and the phonon density of states.

Publications

- Jichen Feng, Ethan Abraham, Joseph Subotnik, and Abraham Nitzan. "Nuclear Angular Momentum Generation in Thermally Driven Chiral Systems." arXiv preprint arXiv:2508.06738(2025). Accepted by PRB. DOI: https://doi.org/10.1103/zbzn-t9bv
- Jichen Feng, Ethan Abraham, Joseph Subotnik, and Abraham Nitzan. "Rectification of Vibrational Energy Transfer in Driven Chiral Molecules." The Journal of Chemical Physics 163, no. 23 (2025).

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