Chiral asymmetry in elastic scattering of vortex electrons by molecules

Valentina Kolovertnova Kirill V. Bazarov
Oleg I. Tolstikhin

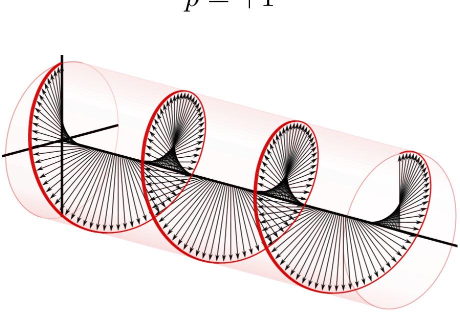
Moscow Institute of Physics and Technology (MIPT)

Chiral asymmetry

Chiral properties of objects manifest themselves only when interacting with a chiral probe.

Example: Circularly polarized photon is a chiral probe in photoelectron circular dichroism (PECD):

$p = +1$ $\qquad$ $p = -1$



Vortex states

$$(-\frac{1}{2}\Delta - E_\nu)\psi_\nu(\mathbf{r}) = 0$$

plane-wave states

fixed momentum

$$\psi_{\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{kr}}$$

$$\nu = \mathbf{k}$$

vortex states

fixed projection of angular momentum on vortex axis

$$\psi_{mq\theta_q}(\mathbf{r}) = J_{|m|}(q_\perp r_\perp)e^{im\varphi_r + iq_z z}$$

$$\nu = m, q, \theta_q$$

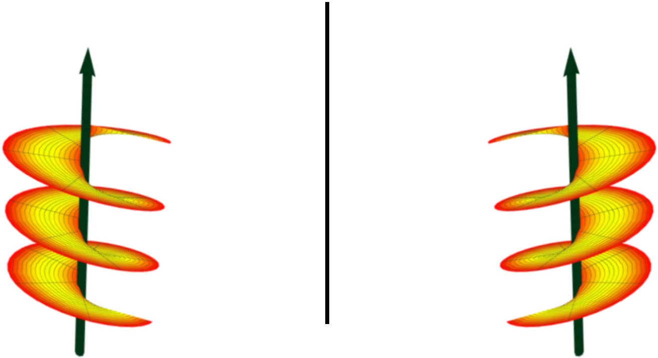
$$q_\perp = q\sin\theta_q,\ q_z = q\cos\theta_q$$

$J_{|m|}$ - Bessel functions of the first kind

Vortex states

Vortex electrons are chiral objects:

$J_{|m|}(q_\perp r_\perp) e^{iq_z z + im\varphi_r}$ $J_{|m|}(q_\perp r_\perp) e^{iq_z z - im\varphi_r}$



a vortex electron can serve as a chiral probe

We analyze the scattering of electrons by chiral molecules, which is governed by the Schrödinger equation:

$$(-\frac{1}{2}\Delta + V(\mathbf{r}) - E_\nu)\psi_\nu(\mathbf{r}) = 0 \quad (1)$$

$V(\mathbf{r})$ - finite-range potential

1. plane-wave scattering states:

$$\psi_{\mathbf{k}}(\mathbf{r})|_{r\to\infty} = \psi_{\mathbf{k}}(\mathbf{r}) + f_{\mathbf{k}}(\Omega_r)\frac{e^{ikr}}{r}$$

2. vortex scattering states:

$$\psi_{mq\theta_q}(\mathbf{r})|_{r\to\infty} = J_{|m|}(q_\perp r_\perp)e^{iq_z z + im\varphi_r} + f_{mq\theta_q}(\Omega_r)\frac{e^{iqr}}{r}$$

Observables

Plane-wave case:

1. total cross section:

$$\sigma_{\mathbf{k}} = \int |f_{\mathbf{k}}(\Omega_r)|^2 d\Omega_r \tag{2}$$

2. total cross section averaged over the direction of **k**:

$$\sigma_k = \int \sigma_{\mathbf{k}} \frac{d\Omega_k}{4\pi} \tag{3}$$

3. Instead of differential cross section, we use angular distribution of scattered electrons:

$$w_{\mathbf{k}}(\Omega_r) = \frac{1}{\sigma_k} |f_{\mathbf{k}}(\Omega_r)|^2 \tag{4}$$

Observables

Vortex case:
Similar to (4):

$$w_{mq\theta_q}(\Omega_r) = \frac{1}{\sigma_q}|f_{mq\theta_q}(\Omega_r)|^2 \tag{5}$$

ZRP

The molecular zero-range potential (ZRP) model: interaction between the incident electron and N-atom target molecule is represented by N ZRPs located at the nuclei

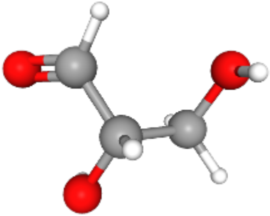


Figure: Glyceraldehyde molecule ($C_3H_6O_3$) is a chiral target

$$V(\mathbf{r}) = \sum_{i=1}^{N} V_{\mathrm{ZRP}}(\mathbf{r} - \mathbf{R}_i; \varkappa_i)$$

$$V_{\mathrm{ZRP}}(\mathbf{r}; \varkappa) = \frac{2\pi}{\varkappa}\delta(\mathbf{r})\frac{\partial}{\partial r}r \qquad (6)$$

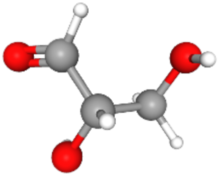
$\mathbf{R}_i$ – position of the i-th atom

$\varkappa_i$ – depth of the i-th potential well

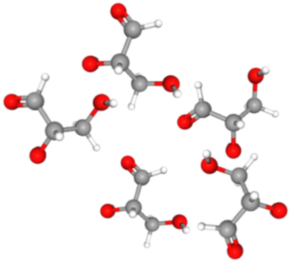
The main results

single molecule

cloud of randomly oriented molecules



$\longrightarrow$



observables averaged over molecule orientations:

$$\langle \mathcal{O} \rangle \equiv \int \mathcal{O}(\omega_{\mathrm{MF}}) \, \frac{d\omega_{\mathrm{MF}}}{8\pi^2}, \qquad (7)$$

The main results

$$\boxed{\left\langle w_{mq\theta_q}(\Omega_r)\right\rangle = \frac{1}{4\pi}\sum_{L=0}^{\infty} b_L^m(\theta_q) P_L(\cos\theta_r)} \tag{8}$$

where

$$b_L^m(\theta_q) = \frac{1}{2\pi(2L+1)\sigma_q}\sum_{JM\lambda\mu}\sum_{\bar{J}\bar{M}\bar{\lambda}\bar{\mu}} f_{JM}^{\lambda\mu} f_{\bar{J}\bar{M}}^{\bar{\lambda}\bar{\mu}*}\Theta_{\lambda|m|}(\cos\theta_q)\Theta_{\bar{\lambda}|m|}(\cos\theta_q)$$
$$\times(-1)^{m+\mu+M}\sqrt{(2J+1)(2\bar{J}+1)}C_{\lambda-m\bar{\lambda}m}^{L0} C_{\lambda-\mu\bar{\lambda}\bar{\mu}}^{L\bar{\mu}-\mu} C_{J0\bar{J}0}^{L0} C_{JM\bar{J}-\bar{M}}^{L\mu-\bar{\mu}} \tag{9}$$

$f_{JM}^{\lambda\mu}$ coefficients are defined by the expansion

$$f_{\mathbf{k}}(\Omega_r') = \sum_{JM\lambda\mu} f_{JM}^{\lambda\mu} Y_{\lambda\mu}^*(\Omega_k) Y_{JM}(\Omega_r) \tag{10}$$

and are a property of the molecule. In ZRP model they are given by

$$f_{JM}^{\lambda\mu} = -i^{\lambda-J} 16\pi^2 \sum_{i,j=1}^{N} j_J(kR_i) Y_{JM}^*(\Omega_{R_i}) M_{ij}^{-1}(k) j_\lambda(kR_j) Y_{\lambda\mu}(\Omega_{R_j}). \tag{11}$$

For enantiomer there is a relation

$$\tilde{f}_{JM}^{\lambda\mu} = (-1)^{\mu+M} f_{J-M}^{\lambda-\mu} \tag{12}$$

1. it can be seen that the coefficients b_L^m have the properties

$$\tilde{b}_L^m(\theta_q) = b_L^{-m}(\theta_q) \tag{13}$$

$$b_0^m(\theta_q) = b_0^{-m}(\theta_q) = \tilde{b}_0^m(\theta_q). \tag{14}$$

   changing the handedness of the molecule $\equiv$ changing the handedness of the incident vortex electron.
2. for a chiral molecule $\tilde{b}_{L>0}^m(\theta_q) \neq b_{L>0}^m(\theta_q) \Rightarrow$, so changing the enantiomer or the sign of m results in a change of the distribution.

Thus, b_L^m with $m \neq 0$ are **chiral-sensitive**.

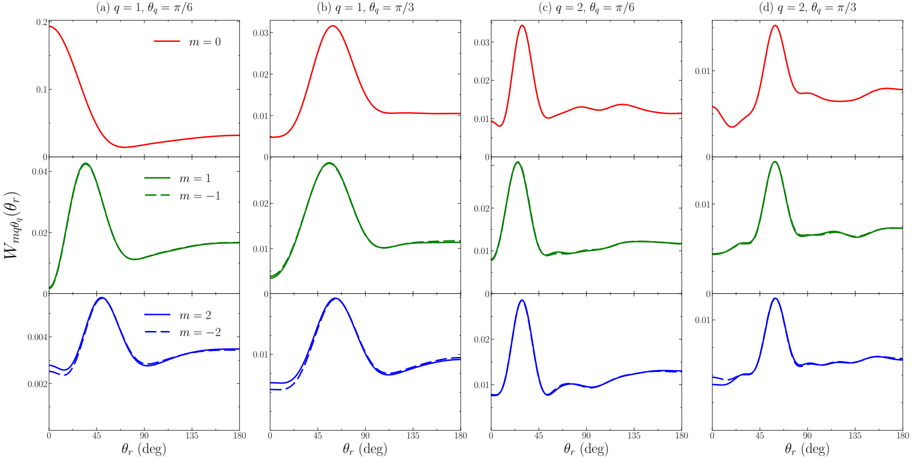
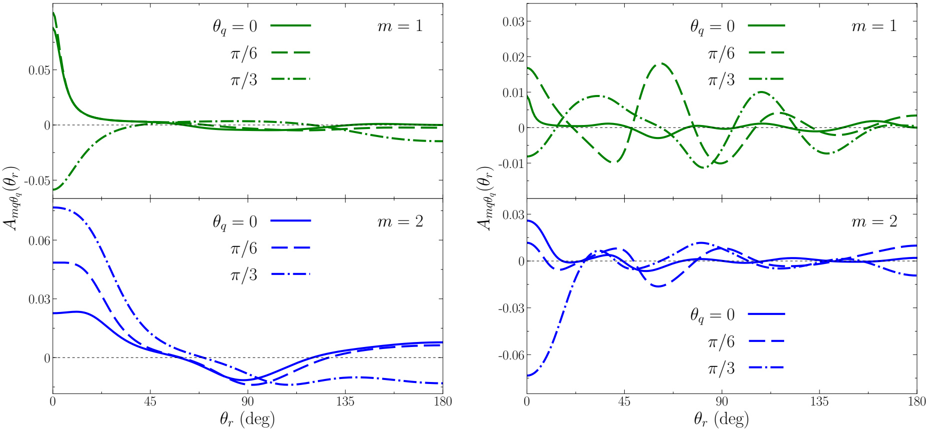


Figure: Angular distributions in the vortex case averaged over molecular orientations. The dashed lines showing results for $m < 0$ are close to but do not coincide with the corresponding solid lines for $m > 0$

To highlight the difference, consider the asymmetry of the distribution:

$$A_{mq\theta_q}(\theta_r) = \frac{W_{|m|q\theta_q}(\theta_r) - W_{-|m|q\theta_q}(\theta_r)}{W_{|m|q\theta_q}(\theta_r) + W_{-|m|q\theta_q}(\theta_r)} \quad (15)$$



The asymmetry of the angular distributions for $q = 1$ and $q = 2$ as a function of the scattering angle θ_r. The solid line shows the asymmetry in the paraxial limit.

We have predicted a new type of chiral asymmetry observable in the elastic scattering of vortex electrons by chiral molecules, which is shown to lead to a measurable asymmetry. This establishes its observability in a typical scattering experiment and potentially opens a new method of probing molecular chirality.

Thank you for the attention!

This work was supported by the Russian Science Foundation (Grant No. 24-12-00055).