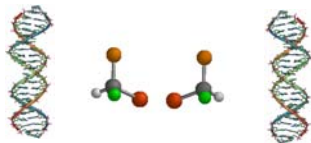


Chiral Molecules

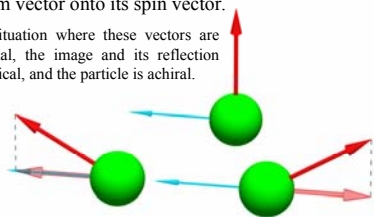
Something is chiral if it cannot be superimposed onto its mirror image. We work with molecules that exhibit this property. The mirror image of a chiral molecule (its *enantiomer*) and the molecule itself exhibit identical chemical behavior except when they interact with other chiral objects.



Chirality of Particles

The handedness of a particle depends on the projection of its momentum vector onto its spin vector.

In the situation where these vectors are orthogonal, the image and its reflection are identical, and the particle is achiral.



If these are parallel, we call the particle right handed.

In the mirror image these vectors are antiparallel; we call the particle left-handed.

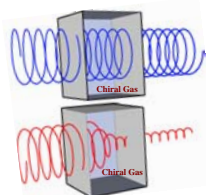
According to the above requirements, photons and electrons can both, in some situations, be chiral objects.

In terms of chirality, electrons are analogous to photons. This analogy is summarized in the following table, which compares the pure states of polarized light and electrons, as well as the corresponding Stokes vectors and density matrices. It is evident from the table that in order to be chiral, electrons must have a longitudinal spin projection, and light must be circularly polarized.

$\begin{pmatrix} 1 \\ 1 \\ 0 \\ 0 \end{pmatrix}$		$\rho = \frac{1}{2} \begin{pmatrix} 1 & \pm i \\ 0 & 0 \\ 0 & 0 \\ 0 & 0 \end{pmatrix}$
$\begin{pmatrix} 1 \\ 0 \\ 1 \\ 0 \end{pmatrix}$		$\rho = \frac{1}{2} \begin{pmatrix} 1 & 0 \\ 0 & \mp 1 \\ 0 & 0 \\ 0 & 0 \end{pmatrix}$
$\begin{pmatrix} 1 \\ 0 \\ 0 \\ 1 \end{pmatrix}$		$\rho = \begin{pmatrix} 1 & 0 \\ 0 & 0 \\ 0 & 0 \\ 0 & 0 \end{pmatrix}$
$\begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}$		$\rho = \begin{pmatrix} 0 & 0 \\ 0 & 1 \\ 0 & 0 \\ 0 & 0 \end{pmatrix}$

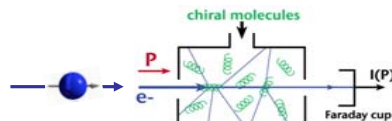
Circular Dichroism

When chiral objects interact with other chiral objects, interesting effects occur. For example, when circularly polarized light interacts with chiral gases, one handedness of the light is preferentially absorbed. This effect is called circular dichroism.



Electron Circular Dichroism

Given the discussed analogy between electrons and photons, we expect that an effect similar to circular dichroism would occur with coherent electron sources and chiral molecules. We have developed the following experiment to measure this effect:



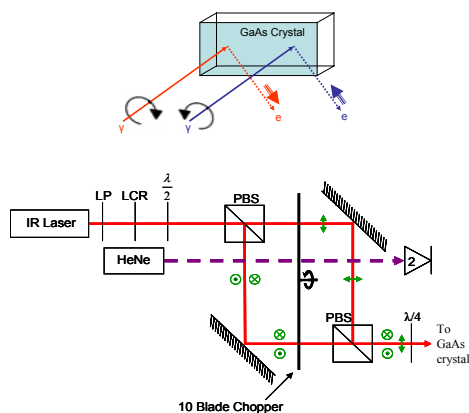
A beam of longitudinally polarized electrons passes through a chiral vapor target. The helicity of the beam is flipped rapidly. Electrons are absorbed according to their spin, and the resulting current is measured on a Faraday cup.

$$A = \frac{I^+ - I^-}{I^+ + I^-}$$

A unitless quantity called Asymmetry quantifies this difference in absorption. I^+ and I^- are the amounts of forward and backward polarized electrons which traverse the target. If $A=0$, there is no effect. However, if $|A|>0$, electron circular dichroism is occurring.

Creation of Polarized Electron Beams

The semiconductor gallium arsenide (GaAs) emits polarized electrons when illuminated with circularly polarized light. The spin of the electrons depends on the helicity of the light.

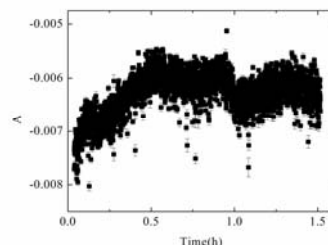


The apparatus above is designed to flip rapidly between RCP and LCP light. A laser beam is split into two arms of opposite polarization by a polarizing beamsplitter. (The polarization of each beam is indicated by the green arrows.) A second Polarizing beamsplitter redirects the beams so that they both point at the crystal. A chopper rotates so that only one polarization reaches the crystal at a time. The elements before the first cube (a polarizer, liquid crystal retarder, and half-waveplate) may be adjusted to equalize the intensities of the two beams of light which traverse the apparatus. For a given point on the crystal, the emission current produced is directly proportional to the intensity of the light which traverses this apparatus.

Problems

- A is predicted to be very small (on the order of 10^{-4}) [1,2].
- Any helicity-dependent intensity or spatial variation of the light incident on the GaAs will mimic a real asymmetry.
- The observed optical **instrumental asymmetry** (any asymmetry that is not due to chiral effects) is large and exhibits long-term drift.

To measure instrumental asymmetry we send electron beams with rapidly flipping spins through an achiral gas. In this case A should be zero, but it usually is not. The following figure is an example of typical optical instrumental asymmetry drift.



The only solution is to force the instrumental asymmetry to zero using external means.

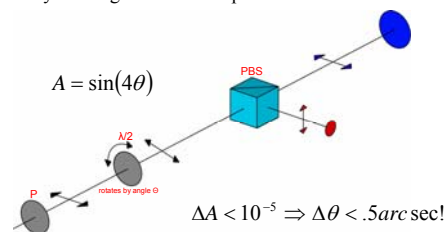
Controlling Asymmetry

We are able to control the optical instrumental asymmetry, which we define as

$$A' = \frac{X^+ - X^-}{X^+ + X^-}$$

where X^+ and X^- are now the intensities of the RCP and LCP light. This intensity asymmetry is directly correlated with the current asymmetry.

In order to force the intensity asymmetry to zero, the intensities of the two beams in our apparatus must be the same. This can be done by rotating the half waveplate.

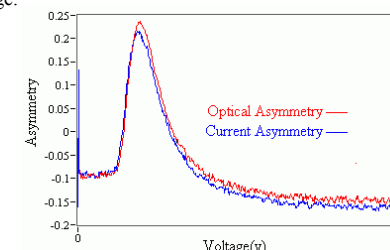


$$\Delta A < 10^{-5} \Rightarrow \Delta \theta < .5 \text{ arc sec!}$$

Since the asymmetry drifts over time, this process must be automated. However, this cannot be done by electronically rotating the half-waveplate, because in order to tune the asymmetry in steps of 10^{-5} , a rotation precision of fractions of an arcsecond is required. This is too close to the limit of what today's nanorotators can produce. Instead, we fine-tune the intensity asymmetry using a liquid crystal variable retarder. This device varies its retardance as a function of a computer controllable voltage, which has the effect of slightly redistributing intensity amongst the two beams.

Active Feedback

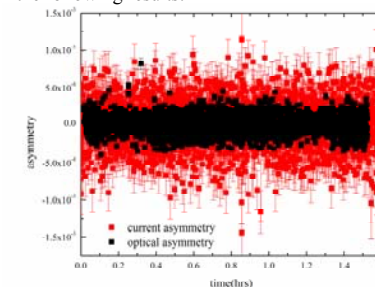
Once we are able to control the asymmetry via computer, we can feed back on any asymmetry we measure and use the LCR to force the asymmetry back to zero. The following curve describes the optical and current asymmetry as a function of voltage.



Once this curve is obtained, we can use a linear approximation about $A=0$ to feed back on nonzero asymmetries. To do this we take an asymmetry measurement and output a voltage according to

$$V_{\text{out}} = -k^{-1}A + V_{\text{previous}}$$

where V_{out} is the output voltage, k is the slope of the curve at $A=0$, and V_{previous} is the voltage output from the previous iteration of the feedback algorithm. Using this algorithm we obtain the following results:



When no feedback is used to force the asymmetry to zero, it drifts by as much as .015 (100 times greater than the asymmetry we would like to measure) in less than a day. However, when we apply feedback, the asymmetry remains at $3.7 \times 10^{-6} \pm 2.2 \times 10^{-6}$ for over 1.5 hours. The active feedback method is successful at maintaining asymmetry at acceptable levels, and can be further improved by increasing feedback resolution, and maintaining temperature stability of the optical system. We expect that employing this system will allow us to make accurately measure a true electron circular dichroism asymmetry.

This research was supported by a grant from UCARE and the National Science Foundation Grant No. PHY - 0653379

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