A Search for the Origins of Biological Homochirality

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(May 17, 2000)

Ever since Pasteur's discovery more than 150 years ago of the handedness of the amino acid asparagine [1], it has been known that nearly all organic molecules associated with natural living matter are chiral. Because he identified such molecules by their propensity to rotate polarized light, the enantiomers (mirror images) have historically been classified as either L- or D-type, corresponding to levo (left) or dextro (right) rotations. Of the 20 amino acids which comprise all proteins found in nature, all but one (glycine) is L-type. This ultimately leads to the characteristic right-handed twist of the DNA helix.

Because chemical interactions of biomolecules are a result of parity-conserving electromagnetic interactions, it is now generally presumed that the origin of biological homochirality is abiotic [2]. That is, life could not have occurred until after some global symmetry breaking produced a pool of homogenous chiral molecules. Very shortly after the discovery of parity nonconservation in nuclear β decay [3], Vester [4] proposed a mechanism exploiting the circularly polarized bremsstrahlung produced by β-decay electrons [5] to produce an enantiomeric excess by either synthesis or degradation. This hypothesis is particularly appealing because it leads to the right sign: The antiparallel electrons produced in β decay lead to left circularly polarized bremsstrahlung which in turn can produce an excess synthesis of L-proteins.

In the 70's, a group at Stanford initially reported evidence for preferential degradation of D(L)-leucine by antiparallel (parallel) longitudinally polarized electrons (120 keV) [6]. However, since that time, several workers (including the original authors) have failed to reproduce those results [7,8]. The Stanford group suggested this might be attributable to the dependence of the bremsstrahlung spectrum on the irreproducible geometry [9]. Unfortunately, because the effect is so weak, there has never been any more definitive experimental evidence to confirm this hypothesis. Quite recently, there have been reports of another positive effect in which the impact of β-decay electrons produced an excess of R-sodium chlorate in a racemic solution (equal L- and R-enantiomer populations) while positrons produced the L-enantiomer [10] and this has reopened this old idea.

The accelerator system that is being proposed for RIA offers a unique opportunity for us to address this nagging question by taking advantage of the capabilities of the booster to accelerate very high mass to charge ratio ions from rest to reduced energies in excess of 500 keV/amu. At such velocities, it is possible to exploit the technique of Coulomb Explosion Imaging (CEI) [11] to measure the absolute chirality of individual molecules. Furthermore, because of the flexibility of the injector layout it will be possible to employ an ion source on an open-air high voltage platform thus permitting the use of circularly polarized photons for photoionization and longitudinally polarized electrons for electron impact ionization.

There are several small (< 100 amu) organic molecules, thought to be prevalent in the chain of chemical evolution preceding the origin of life [12], which can be synthesized as chiral molecular ions for acceleration and analysis by CEI. In particular, consider the case of the ethylene molecular ion. Neutral ethylene (H_2CCH_2)is planar in the ground state and hence achiral. However, there is a growing body of evidence to suggest that the ground state geometry of the molecular ion is twisted [13] and hence chiral. Thus, the ionization of the racemic neutral species by either photon or electron impact could produce a chiral daughter ion. This is in sharp contrast to all previous measurements which have sought to *degrade* a racemic mixture. In this experiment we will use both polarized photons and polarized electrons to *produce* a chiral molecule.

Because CEI experiments are 100% efficient, the rate limit in the experiment is not the ion source or accelerator efficiency but rather detector count rates. Using the large area wire chambers that have been employed in the past [14,15], this would lead to a practical limit of ~ 1 kHz. Thus, in a 30 hour period, we could analyze 10^8 chiral molecules yielding a sensitivity of 100 ppm for chiral discrimination — more than 2 orders of magnitude improvement over previous measurements. Such an experiment would represent a dramatic achievement and would provide definitive evidence either for or against the Vester hypothesis.

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