

Remote Detection of Chiral Organics on Surfaces Using Second-Order Nonlinear Optics. D.P. Winebrenner^{1,2}, W.E. Asher, and E. Willard-Schmoe¹, ¹Applied Physics Laboratory, Box 355640, University of Washington, Seattle, WA 98195, dpw@apl.washington.edu, ²Dept. of Earth and Space Sciences, University of Washington, Seattle, WA 98195.

Introduction: Homochirality, or even large enantiomeric excesses in amino acids, sugars, or larger molecules formed from those constituents (e.g. proteins) are generally considered to be signs of biogenic origin.

Detection of enantiomeric excesses by optical means may facilitate remote methods for targeting in situ sample collection, or for placing sample collection sites in context. Elastic-scattering (i.e., linear) optical methods based on circular dichroism spectroscopy or differential scattering of orthogonal circular polarizations typically require very high abundances and enantiomeric excesses to produce a small signal (see, for example, [1]).

By contrast, methods based on second-order nonlinear optical spectroscopy display sensitivity to abundances of chiral molecules several orders of magnitude larger than linear methods [2]. Moreover, such methods respond specifically to chiral molecules at interfaces, rather than in bulk materials – this is of special interest because of the tendency for microbes to be found on surfaces in nature.

Toward Detection Limits: The existing literature on nonlinear second order methods to probe chiral molecules at interfaces is concerned primarily with the derivation of information on molecular structure, rather than on detection limits.

Here we reanalyze published data on chiral second-harmonic generation involving organic molecules on surfaces to derive estimates of sensitivity of detection which might be expected. We compare those estimated sensitivities with observed detection sensitivities for non-organic compounds containing strongly polarizable (i.e., nonlinearly active) bonds which we have established using the related method of sum-frequency generation.

References: [1] Sparks, W.B. et al., Proceedings of the National Academy of Sciences, www.pnas.org/cgi/doi/10.1073/pnas.0810215106, 2009. [2] Simpson, G.J., ChemPhysChem 5, 1301 – 1310 DOI: 10.1002/cphc.200300959, 2004.