

Chiral plasmons & orientation dependent handedness

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Chirality and handedness are universal asymmetries with manifestations such as the homochirality of life on earth, the arrow of time, and handedness of light in vacuum. Lord Kelvin's definition of chirality as the lack of mirror symmetry [1], which has guided thinking for over a century, is based on the discrete measure of the reflection transformation. Yet inspection of chiral structures such as shoes and potatoes suggests the necessity for a continuous measure of handedness [2], which is formally recognized as the topological connectedness of chirality [3]. Accordingly, a chiral structure need not be handed, and an achiral structure may show handed response upon measurement. We illustrate these concepts experimentally, through measurements of Raman optical activity on a gold nanosphere dimer. We show the rather counter-intuitive effects of orientation dependent handedness of a nominally achiral structure, and passage from right- to left-handed response without crossing an achiral state. The experimental system can be identified as the fundamental building block of chiroptical response; namely, a uniaxial structure that sustains collinear magnetic and electric dipole as suggested by P. Curie [4], which in this case is driven by chiral plasmons.

Beside their giant Raman optical activity, chiral plasmons open up the magnetic channel in light-matter interactions, by serving as magnetic nanoantennas that operate at optical frequencies. Otherwise, we principally see through the electric component of light. The effective excitation of magnetic plasmons on nonmagnetic materials such as gold is strictly a shape effect, due to the breakdown of the long-wave limit. On nanostructures with features that reach the wavelength of the excitation resonance, electric, magnetic and quadrupolar forces exerted by light become comparable. This leads to metamaterials with unusual optical properties that are used in applications such as the generation of left-handed light. In this regard, plasmonic nanosphere dimers constitute prototypical metamaterials, with rich optical response that can be understood by noting that they are the nanoanalog of diatomic molecules.

- [1] *Baltimore Lectures on Molecular Dynamics and the Wave Theory of Light*, Lord Kelvin, (Cambridge Library Collection, 1904).
- [2] R. B. King, *The Ruch "Shoe-Potato" Dichotomy in the Right-Left Classification Problem*, Ann. N.Y. Acad. Sci. 988, 158 (2003).
- [3] E. Efrati and W. T. M. Irvine, *Orientation dependent handedness and chiral design*, Phys. Rev. X, 4, 011003 (2014).
- [4] P. Curie, *Sur la symmétrie dans les phénomènes physique symmétrie d'un champ électrique et d'un champs magnétique*, J. Physique, 3, 393 (1894).