Coherent Chiroptical Spectroscopy and Quantum Interference

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In chiroptical spectroscopy, there have been quite extensive efforts to make an enhanced detection of weak optical activity signal from chiral molecules in isotropic media using self- and active-heterodyne detection schemes. In particular, a significant amplification effect of weak chiroptical signal was shown to be achievable by using ellipsometric detection with employing a cross polarization detection scheme. This is one of the self-heterodyne measurement methods because the interference term between the incident beam and chiroptical free induction decay field is detected. An alternative approach is to use an independently propagating radiation to achieve an active heterodyne detection of weak chiroptical signal. Nonetheless, the common element in both the active- and self-heterodyne-detected chiroptical measurements is the cross polarization geometry, which has also been used to demonstrate weak value amplification in quantum optics and metrology. The cross polarization method has also been shown to be useful for developing multi-dimensional chiroptical spectroscopy, which is a combination of chiroptical and coherent multi-dimensional spectroscopic techniques. The rotationally non-invariant elements of the fourth-rank tensors that are associated with two-dimensional electronic spectroscopy contain information on nonlinear chiroptical responses from chiral molecules in condensed phases. Only recently, its experimental feasibility has been demonstrated for light-harvesting complex proteins. Also, here it will be discussed that an improved optical enantioselectivity, or related dissymmetry factor, of chiral molecules can be achieved by making the energy density of electric field component small as well as that of magnetic field component large. In particular, we have shown that a negative-index metamaterial can be of use to enhance local optical chirality, which is a measure of chirality of light not material, in the vicinity of nano-scale cavities. We anticipate that the cross polarization method in combination with properly designed metamaterials would allow us to control local electric and magnetic field energy densities and to detect extremely weak one and even multi-dimensional chiroptical spectroscopy signals in the future.