Biological membranes are actively involved in a multitude of processes in living cells; therefore, a detailed characterization of their structure, dynamics, and function is essential for an understanding of living organisms at the molecular level. In this work, we made use of the high spatial and temporal resolution offered by computer simulations to investigate the behavior of several molecular species which associate with cellular membranes. Using a combination of classical molecular dynamics simulations and ab initio electronic structure calculations, we were able to characterize nonlinear optical properties of membrane-embedded fluorescent probes and thus contribute to establishing two-photon polarization microscopy as a tool of structural biology. Moreover, our molecular dynamics simulations provided an atomistic picture of the reversible membrane binding of recoverin, a neuronal calcium-sensing protein involved in vision adaptation, and they also yielded an important insight into the mechanism of its calcium-induced myristoyl switch. In addition, we examined the biological role of cholesterol oxidation and compared two methods of representing transmembrane voltage in molecular dynamics simulations.