Ab Initio Studies of Nonlinear Optical Responses of Magnetic Semiconductors

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Second-order nonlinear optical (NLO) responses, such as second harmonic generation (SHG) and bulk photovoltaic effects (BPVE), occur only in noncentrosymmetric materials. These NLO processes have played important roles in materials characterization and also have found applications in optoelectronics, efficient solar energy harvesting and sensitive terahertz radiation detection. Until recently, studies of second-order NLO responses have been focused on nonmagnetic materials with broken spatial inversion symmetry such as few-layer transition metal dichalcogenide semiconductors (see, e.g., [1-2]) and BN nanotubes (see, e.g., [3-4]) as well as quasi-one-dimensional trigonal tellurium and selenium [5]. However, magnetism not only can break the spatial inversion (*P*) symmetry of centrosymmetric crystals but also introduce additional NLO processes in materials with already broken *P*-symmetry by breaking time-reversal (*T*) symmetry, thus allowing us to control light-matter interactions by, e.g., magnetic field. We have recently developed *ab initio* computational methods for calculating magnetism-induced second-order NLO properties and also applied them to study a number of magnetic materials including topological semimetals [6-7]. Moreover, we have shown that NLO responses are a powerful probe of geometric structure of quantum states in the materials [6-7]. In this talk, I will present the salient results of our *ab initio* calculations for several magnetic semiconductors including *PT* symmetric antiferromagnetic CrI_3 bilayer [8], Cr_2O_3, $MnTiO_3$ and $MnPS_3$ as well as antiferromagnetic relativistic semiconductor $Fe_2Mo_3O_8$ [9] and ferrimagnetic $Eu_2MnSi_2O_7$ [10].

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