Spin relaxation, dephasing and diffusion in solids from ab-initio density-matrix dynamics

Yuan Ping[1,2]

1. University of Wisconsin, Madison
2. University of California, Santa Cruz

Spin relaxation, dephasing and diffusion are at the heart of spin-based information technology. Accurate theoretical approaches to simulate spin lifetimes (τ_s), i.e. how fast the spin polarization and phase information will be lost, are important to the understandings of underlying mechanism, and invaluable to search for promising spintronic material candidate. Recently, we develop the first-principles real-time density-matrix (FPDM) approach to simulate spin dynamics for general solid-state systems [1,2]. Through the complete first-principles' descriptions of light-matter interaction and scattering processes including electron-phonon, electron-impurity and electron-electron scatterings with self-consistent spin-orbit coupling, as well as *ab-initio* Land'e *g*-factor[3], our method can predict τ_s, regardless of crystal symmetry, as a function of carrier density and temperature, under electric and magnetic fields[2,3].

By employing this method, we successfully reproduce experimental results of spin lifetime and ultrafast Kerr rotation of disparate materials, and identify the key factors affecting spin relaxation, dephasing, and diffusion[4] in different materials. Specifically, we predict that germanene has long τ_s (~100 ns at 50 K), a giant spin lifetime anisotropy and spin-valley locking effect under electric fields[4], making it advantageous for spin-valleytronic applications. Based on our theoretical derivations and *ab-initio* simulations, we propose a new useful electronic quantity, named spin-flip angle $\theta_{\uparrow\downarrow}$, for the understanding of spin relaxation through intervalley spin-flip scattering processes[5]. We then show how *g* factor fluctuations lead to spin dephasing in halide perovskites under external magnetic field. Our method can be further applied to other emerging materials and extended to simulate exciton spin dynamics and steady-state photocurrents due to photogalvanic effect.

References

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