Spin Relaxation in Solids from Ab Initio Density-matrix Dynamics

Yuan Ping
University of California, Santa Cruz, 1156 High Street, Santa Cruz, USA
yuanping@ucsc.edu

Abstract

Designing new quantum materials with long-lived electron spin states is in urgent need of a general theoretical formalism and computational technique to reliably predict spin lifetimes. We present a new, universal first-principles methodology based on density matrix (DM) dynamics for open quantum systems to calculate the spin relaxation time of solids with arbitrary spin mixing and crystal symmetry. In particular, this method describes contributions of the Elliott-Yafet (EY) and D'yakonov-Perel' (DP) mechanisms to spin relaxation, on an equal footing[1]. Our ab initio predictions are in excellent agreement with experimental spin lifetime for a broad range of materials, such as Si, Fe, MoS_2, graphene as well as GaAs[1,2].

We then discuss our implementation of real-time DM dynamics for ultrafast Kerr rotation and studied spin dynamics under external electric and magnetic field [2]. Through the complete theoretical descriptions of pump, probe and scattering processes including electron-phonon, electron-impurity and electron-electron scattering, our method can directly simulate the ultrafast pump-probe measurements for coupled spin and electron dynamics and is applicable to any temperatures and doping levels. We use this method to simulate spin dynamics of GaAs and WSe_2 and obtain excellent agreement with experiments. It is found that the relative contributions of different scattering mechanisms and phonon modes vary considerably between spin and carrier relaxation processes. We further apply our method to investigate the condition of realizing long spin lifetime and spin-valley locking of 2D Dirac materials such as germanene under electric field [3], and understand substrate effects on spin relaxation in graphene[4], resolving the contribution of electron-phonon coupling and proximity effect on spin-orbit coupling.

References

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[2] J. Xu, A. Habib, R. Sundararaman and Y. Ping, under review, preprint: arXiv: 2012.08711 [cond-mat-mtrl-sci], (2021).
[3] J. Xu, A. Habib, R. Sundararaman and Y. Ping, under review, (2021).
[4] A. Habib, J. Xu, Y. Ping and R. Sundararaman, under review, preprint: arXiv:2012.11550 [cond-mat.mtrl-sci], (2021).

Figures

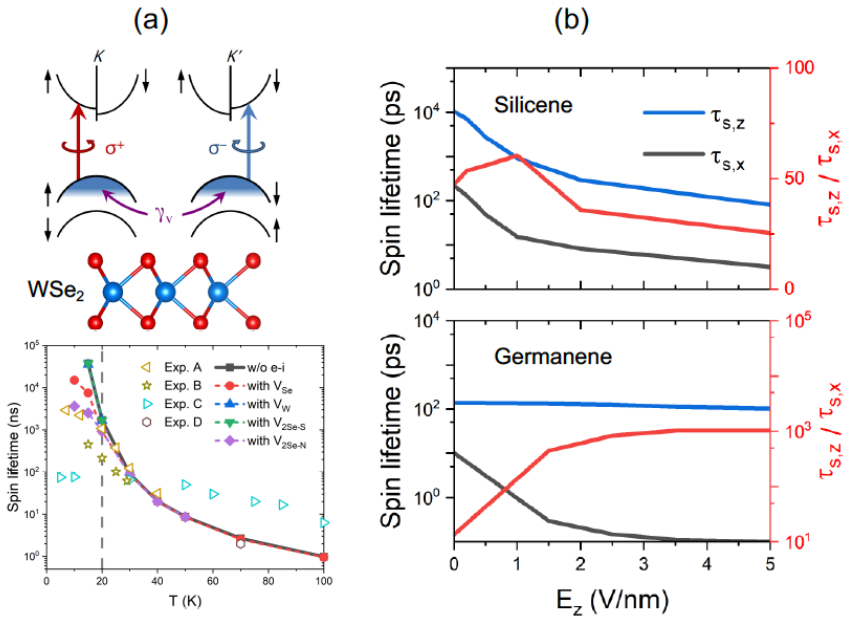

Figure 1: (a)Out-of-plane spin lifetime of WSe_2 with and without point defects (w/o e-i). Certain defects have large effects below 20K[2]. (b) Spin lifetime and its anisotropy of silicene and germanene under electric field. The out-of-plane spin lifetime of germanene is insensitive to applied E field[3].

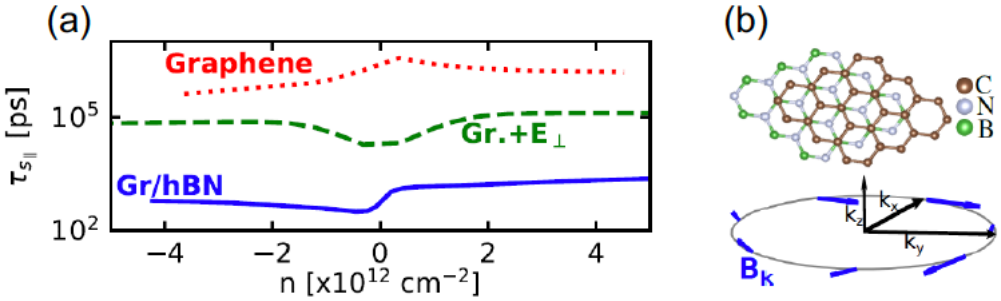

Figure 2: (a) Graphene spin lifetime as a function of carrier density as a single layer (Graphene), under electric field (Gr.+E) and on a substrate (Gr/hBN). (b) Graphene on hBN interface model and its schematic titled internal B field[4].