Spin Relaxation, Dephasing, and Diffusion in Solids from ab-initio Density-Matrix Dynamics

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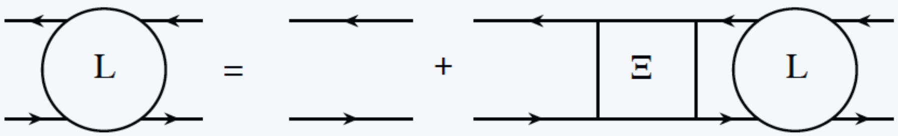
Theory and Computational Method Development

Applications

Poster: Shimin Zhang, "advanced simulations on spin qubits"

Many-Body Perturbation Theory for Excited States[1]



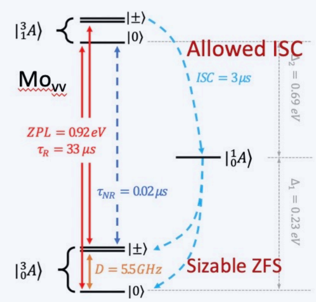
Ab-initio Open Quantum Dynamics[2]

$$i\hbar\dot{\rho} = [H, \rho] + F[\rho]$$

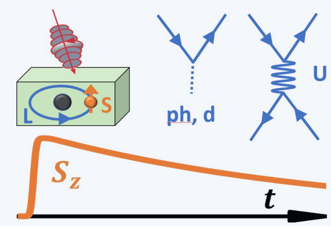
SOC, e-e, e-imp, e-ph, e-photon

DFT **MBPT**

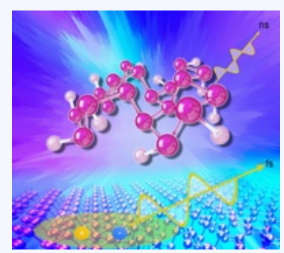
Spin qubits[3]



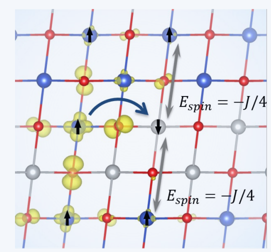
Spin dynamics and transport[2]



Exciton Dynamics[5]



Polaron Transport[6]



Condensed Matter Physics ⟷ **Physical Chemistry**

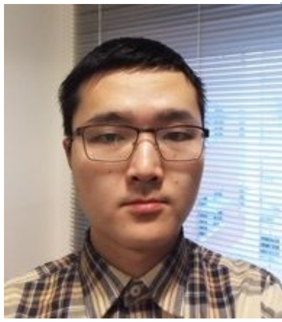
1. Y. Ping et al, *Chem. Soc. Rev.*, 2013. 2. J. Xu, ..Y.P., *Nat. Commun.* 2020. J. Xu, ..Y.P., PRB, 2021. Editor's Suggestions and Highlight: "**A universal model of spin relaxation**".
3. Y. Ping, T. Smart, *Nat. Comput. Sci.* 2021. 4. J. Xu, ..Y.P., *Nano Lett.* 2021; B. Zhao et al, *Nature*, 2021. 5. F. Wu, D. Rocca, Y.P., JMCC, 2019. 6. T. Smart.. Y.P., *npj Comp. Mater.* 2018.

Ping Group

Moving to UW-Madison July 1st 2023

Postdoc and PhD positions are available!

Postdocs:



Feng Wu
(Alibaba QC)

Junqing Xu
(current. Prof. in China)

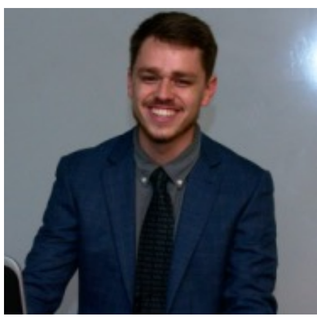
Rafi Ullah

Key theory collaborators:

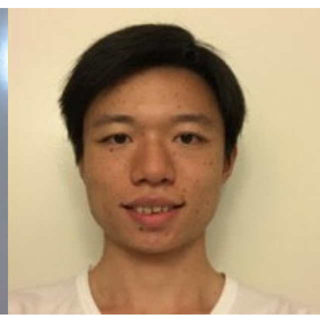
Dario Rocca
(France, U. Lorraine,
now QC Ware)

Ravishankar
Sundararaman
(RPI)

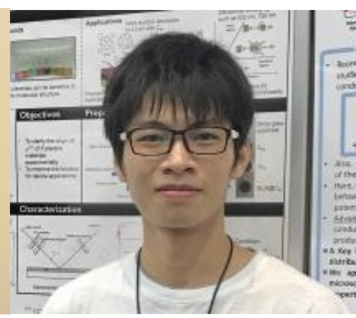
Graduate students:



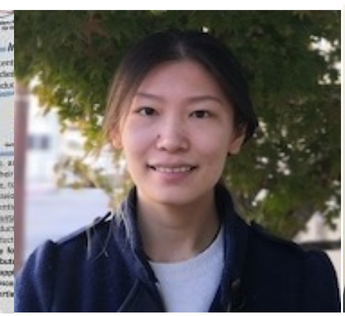
Tyler Smart
(Intel)



Chunhao Guo
(5nd)



Kejun Li
(5nd)



Shimin Zhang
(4nd)

Andrew Grieder
(2st)

http://yuanping.chemistry.ucsc.edu

Outline

- Introduction of spin-based information science
- Theoretical framework for ab-initio density-matrix dynamics in solids
- Spin relaxation and transport in 2D materials
- Spin relaxation and dephasing in halide perovskites
- Conclusion and outlook

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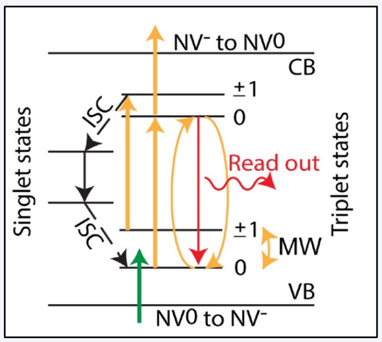
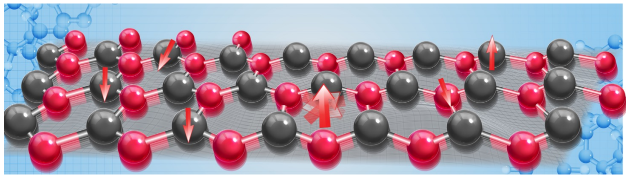
Materials for Quantum Information Science

Critical processes for spin-based quantum information technologies

Optical Readout

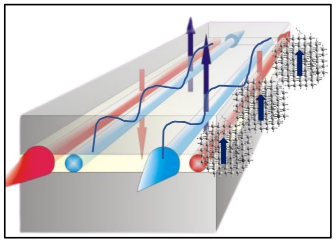


Spin Relaxation and Coherence



(T_1 and T_2, how long the quantum state can survive?)

Quantum Information Transduction



None of the current spin qubit candidates is considered "ideal" for scalable quantum applications

Design new materials for spin qubits in quantum sensing and quantum computing
- Long quantum coherence, efficient readout, quantum transduction

Developed Methodologies for 2D Quantum Defects

Poster: Shimin Zhang, "advanced simulations on spin qubits"

- **Many-body theory –** including screened Coulomb interaction (GW)[1,2,3]
- **Charged cell correction for 2D –** avoid any supercell extrapolation[1,2,4]
- **Exciton radiative recombination –** solving the Bethe-Salpeter Equation (BSE)[3,5,6,7]
- **Nonradiative recombination –** phonon mediated recombination[5,8]
- **Intersystem crossing rate and ODMR –** spin-flip nonradiative transition due to SOC[5]
- **Substrate effect – sum up effective polarizability** [6,11]
- **Quantum Embedding theory for 2D defects (in progress)**

Develop ab initio theory to accurately predict electronic, optical, and dynamical properties for spin defects in 2D materials

$$\Gamma_R(Q) = \frac{4\pi^2 e^2}{\hbar c^2 V}\Omega(Q)^2 \sum_{q,\lambda}\frac{1}{q}\left|\epsilon_{q\lambda}\cdot\langle G|r|S\rangle\right|^2 \times\delta(\Omega(Q)/c-q)$$

$$\Gamma_{NR} = \frac{2\pi}{\hbar} g\left|W_{if}\right|^2 X_{if}(T)$$

$$W_{if} = \left\langle\psi_i(r,R)\left|\frac{\partial H}{\partial Q}\right|\psi_f(r,R)\right\rangle\Big|_{R=R_a}$$

$$X_{if}(T) = \sum_{n,m} p_{in}\left|\langle\phi_{fm}(R)|\Delta Q|\phi_{in}(R)\rangle\right|^2 \times\delta\left(m\hbar\omega_f - n\hbar\omega_i + \Delta E_{if}\right)$$

$$\Gamma_{ISC} = 4\pi\hbar\lambda_\perp^2 \tilde{X}_{if}(T)$$

$$\tilde{X}_{if}(T) = \sum_{n,m} p_{in}\left|\langle\phi_{fm}(R)|\phi_{in}(R)\rangle\right|^2 \times\delta\left(m\hbar\omega_f - n\hbar\omega_i + \Delta E_{if}\right)$$

[1] F. Wu, A. Galatas, R. Sundararaman, D. Rocca, and Y. Ping, Phys. Rev. Mater. 1, 071001(R) (2017).
[2] T. J. Smart, F. Wu, M. Govoni, and Y. Ping, Phys. Rev. Mat. 2, 124002 (2018).
[3] Y. Ping, D. Rocca, and G. Galli, Chem. Soc. Rev. 42, 2437 (2013).
[4] R. Sundararaman, and Y. Ping, J. Chem. Phys., 146, 104109 (2017).
[5] T. J. Smart, K. Li, J. Xu, Y. Ping, NPJ Comput. Mater. 7, 59, (2021).
[6] C. Guo, J. Xu, D. Rocca, and Y. Ping, Phys. Rev. B 102, 205113, (2020).
[7] F. Wu, D. Rocca, and Y. Ping, J. Mater. Chem. C, 7, 12891 (2019).
[8] F. Wu, T. J. Smart, and Y. Ping, Phys. Rev. B, 100, 081407(R) (2019).
[9] Y. Ping and T. J. Smart, Nat. Comput. Sci., 1, 646, (2021)
[10] K. Li, T. J. Smart, Y. Ping, Phys. Rev. Mater (Letter), 6, L042201, (2022)
[11] S. Zhang, K. Li, C. Guo, and Y. Ping, 2D Materials, in press, (2023) arxiv.org/abs/2304.05612

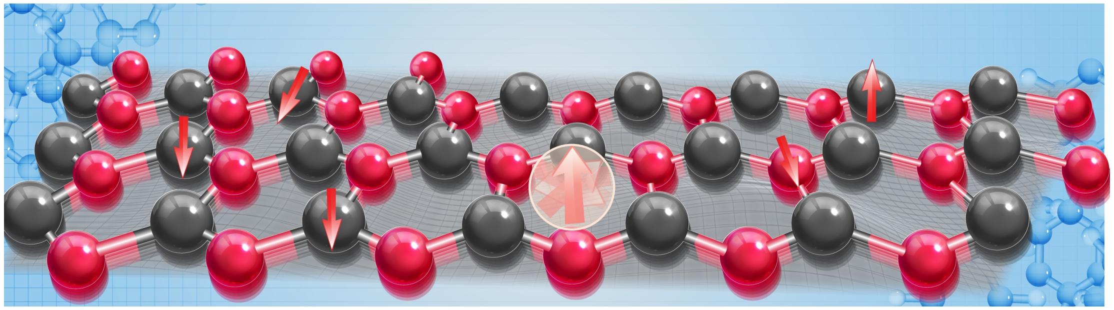
Computational Codes

In-house codes built on top of QuantumESPRESSO and Yambo-code

Quantum Coherence of Spin Qubit

Major Challenges of Quantum Computation:

- **Decoherence** of encoded information – loss of information
- Building large scale fault-tolerant quantum computer

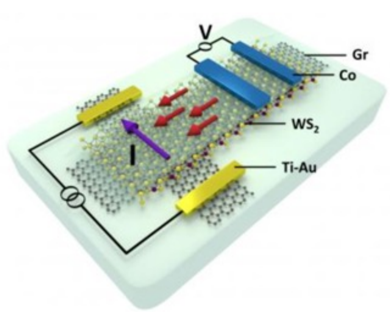


How long the quantum state can survive?
(Localized) spin relaxation T_1 and decoherence T_2

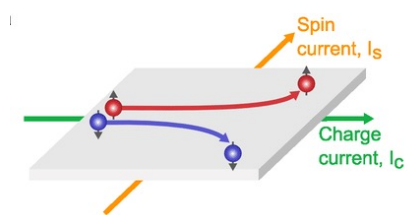
Spin-Based Information Processing

Low-power electronics based on spin manipulation?
(Delocalized spin)

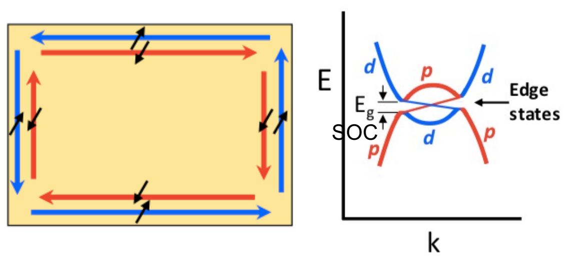
Spin transistors



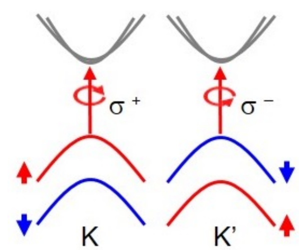
Spin hall effect



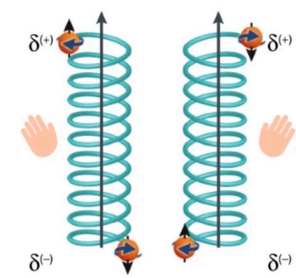
QSH



Valleytronics



CiSS effect



Need spin information transport over long distance

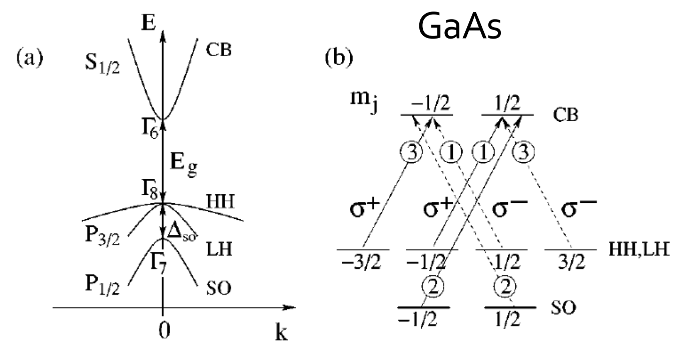
Spin diffusion length² = τ_s . D

Here we use **spin lifetime** (τ_s) including both spin relaxation (T_1) and decoherence (T_2)

Spin Imbalance Generation and Detection in Solids

The generation of spin imbalance:

- Optical excitation with circularly polarized light
- Injection of spin polarization from ferromagnets

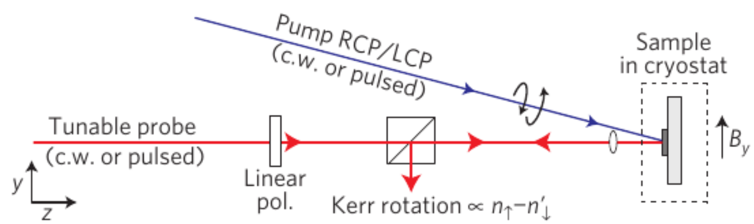


Photon spin (helicity +/-) selectively excites electrons with certain m_j that gives spin polarization

Zutic et al, *Rev. Mod. Phys.* 76, 2, (2004)

The detection of spin relaxation:

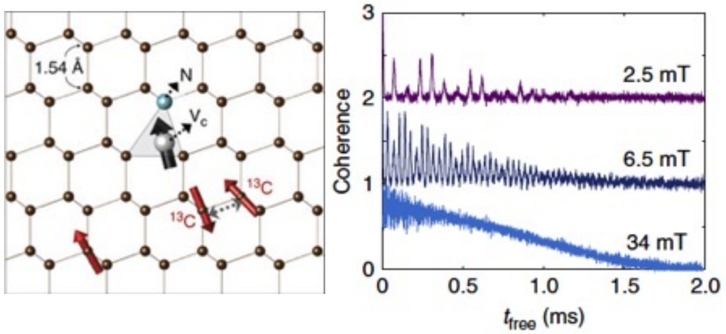
- Time-resolved Kerr rotation
- Spin transport, $L_s{}^2 = \tau_s \cdot D$



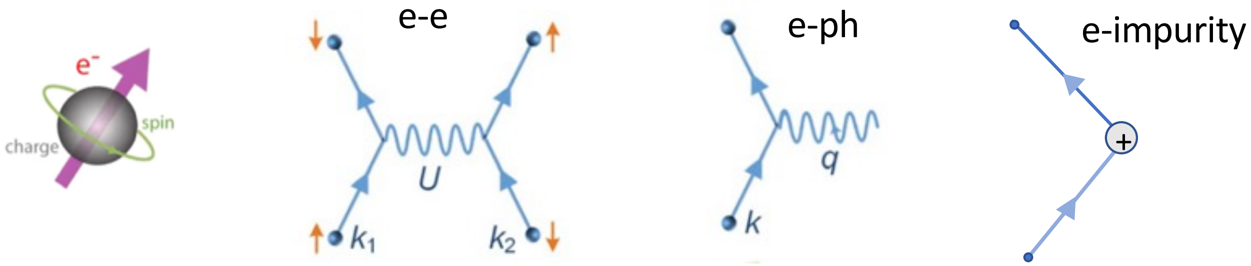
Spin Decoherence and Relaxation Mechanism

- At very low temperature and large B field, decoherence mainly from fluctuated magnetic field by nuclear spin flip-flop transition

- At finite temperature, other effects can be dominant such as phonons, impurities, electron-electron interactions through spin-orbit couplings.

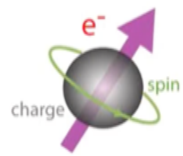


At 20K, NV in diamond spin decoherence time T_2 is ~ms, H. Seo et al *Nat. Commun*. (2016)
SiC divacancy T_2 360 μ_s at 20K and 50 μ_s at 300K

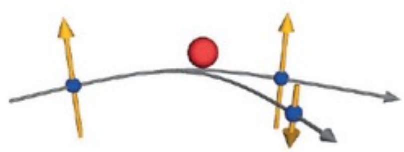


Spin Relaxation and Decoherence Mediated by SOC

Spin-orbit interaction couples spin with phonons, impurities and electrons

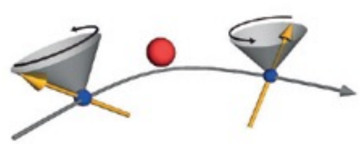


Eilliot Yafet

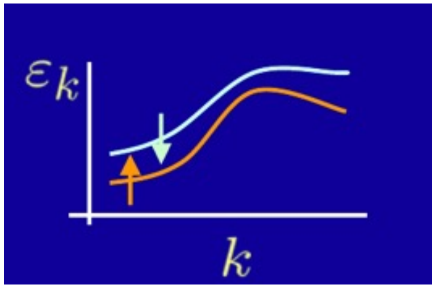


With inversion symmetry

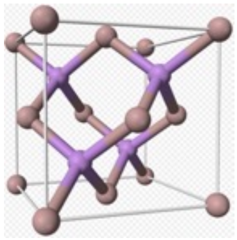
Dyakonov-Perel



Without inversion symmetry



GaAs

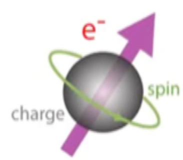


Break spin up/down - Kramers degeneracy in GaAs: $\varepsilon_k^{\uparrow} \neq \varepsilon_k^{\downarrow}$, still has $\varepsilon_k^{\uparrow} = \varepsilon_{-k}^{\downarrow}$

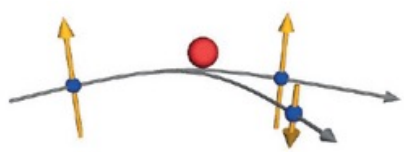
An internal k dependent magnetic field $\Omega(\mathbf{k})$

Spin Relaxation and Decoherence Mediated by SOC

Spin-orbit interaction couples spin with phonons, impurities and electrons

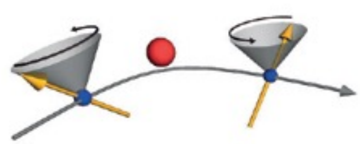


Eilliot Yafet



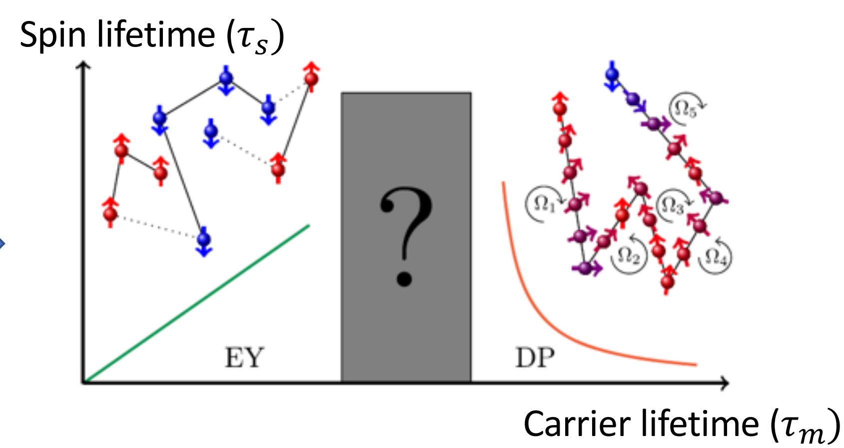
With inversion symmetry

Dyakonov-Perel



Without inversion symmetry

$\boldsymbol{\tau_s \propto \tau_m}$ $\boldsymbol{\tau_s \propto 1/\tau_m}$



$$\tau_s \propto 1\Big/\left(\tau_m.\left(\langle\Omega_{\mathbf{k}}^2\rangle - \langle\Omega_{\alpha}^2\rangle\right)\right);\ \ \Omega_{\mathbf{k},i} = 2\Delta E_{\mathbf{k}}.\, s_{\mathbf{k},i}/\hbar^2$$

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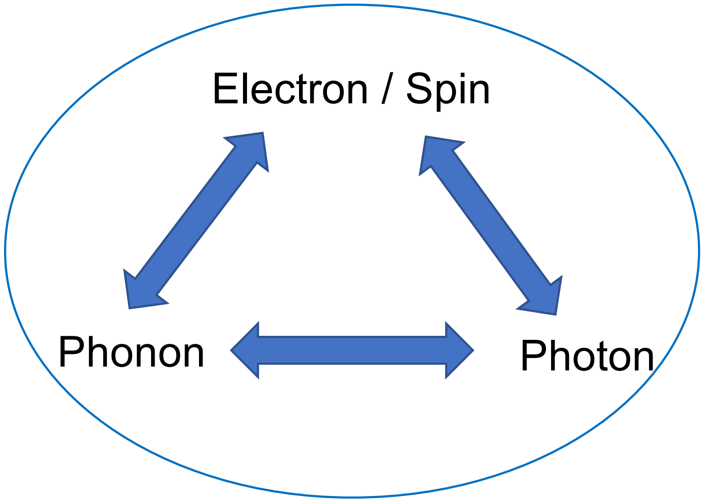
- Introduction of spin-based QIS
- **Theoretical framework for ab-initio spin dynamics in solids**
- Spin relaxation and transport in 2D materials
- Spin relaxation and dephasing in halide perovskites
- Conclusion and outlook

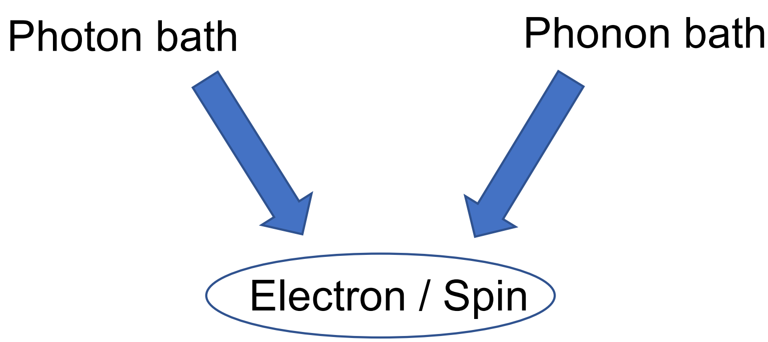
Open Quantum Dynamics

Closed Quantum Systems



Photon, phonon, electron
treated at the equal footing, self-consistently

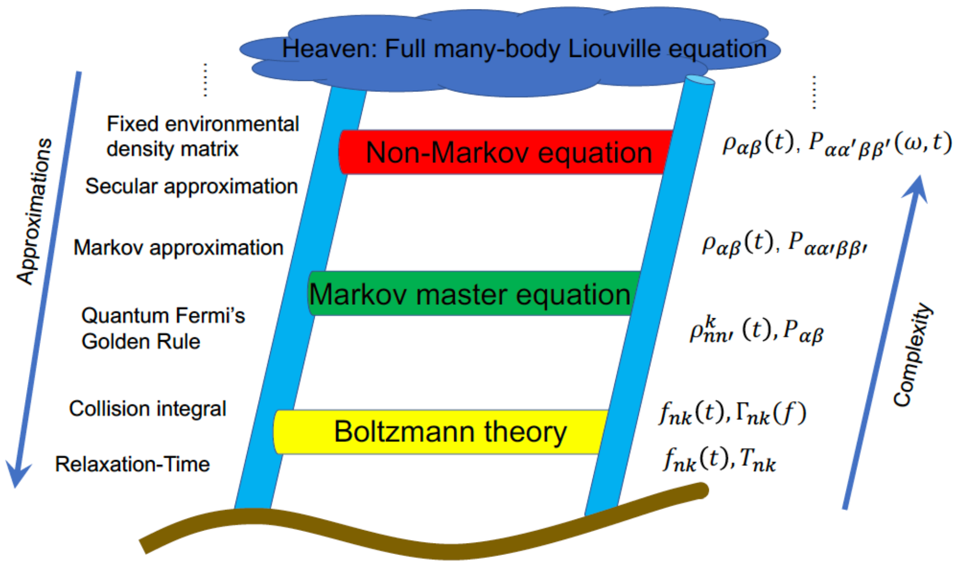
Open Quantum Systems



The bath may or may not have memory
effects depending on the chosen theory

First-Principles Open Quantum Dynamics in Solids

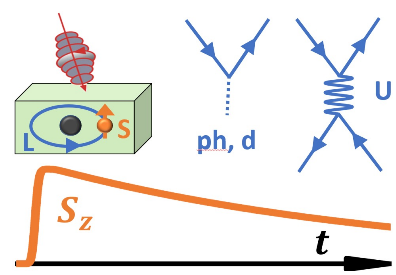
Developed first-principles open quantum dynamics with all decoherence pathways for coupled spin and carrier dynamics for solids



Jacob's Ladder for quantum dynamics

Why we need this theory?

(i) long simulation time (ns-μs)
(ii) adequate for spin dynamics (VS. Boltzmann)
(iii) non-equilibrium (VS. FGR)



"A universal spin relaxation model in solids"
Physics Magazine, 2021

J. Xu et al, Nat. Commun. **11,** 2780 (2020).
J. Xu…Y. Ping, PRB, **104**, 184418, (2021)

Density Matrix Dynamics with Coupled Electron, Phonon and Photon

$$\hat{H} = \hat{H}_0 + \hat{H}^I_{ext} + \hat{H}^I_{scatt}$$

$$\hat{H}_0 = \hat{H}^c_o + \hat{H}^b_o$$ (DFT/MBPT)

$$\hat{H}^I_{ext} = \Delta \sum_{xc}(t) + U(t)$$

$U(t)$: pump or applied field

$\hat{H}^I_{scatt}$: scatterings

$$i\hbar \frac{\partial \tilde{\rho}(t)}{\partial t} = [\tilde{H}^I(t), \tilde{\rho}(t)]$$

Liouville-von Neumann equation in the interaction picture

$$\tilde{\rho}(t) = e^{iH_0 t/\hbar} \rho e^{-iH_0 t/\hbar}$$

$$\tilde{H}^I(t) = e^{iH_0 t/\hbar} \hat{H}^I e^{-iH_0 t/\hbar}$$

$$\frac{\partial \tilde{\rho}(t)}{\partial t} = \frac{\partial \tilde{\rho}(t)}{\partial t}|_{coh} + \frac{\partial \tilde{\rho}(t)}{\partial t}|_{scatt},$$

$$\frac{\partial \tilde{\rho}(t)}{\partial t}|_{coh} = -i[\Delta \sum_{xc}(t) + U(t),\ \tilde{\rho}(t)]$$

$$\frac{\partial \tilde{\rho}(t)}{\partial t}|_{scatt} = \frac{\partial \tilde{\rho}(t)}{\partial t}|_{e-e} + \frac{\partial \tilde{\rho}(t)}{\partial t}|_{e-ph} + \frac{\partial \tilde{\rho}(t)}{\partial t}|_{e-imp}$$

Each scattering term needs scattering matrix elements ($P_{\alpha\alpha',\beta\beta'}$) from first-principles

- Compute observable O in real time dynamics:

  $O = Tr(\tilde{o}\tilde{\rho})$ Spin: $o = S$, Carrier: $O = I$

$$\frac{d\rho_{12}}{dt}|_c = \frac{1}{2}\sum_{345}\left[\begin{array}{c}(I-\rho)_{13} P^c_{32,45}\rho_{45} \\ -(I-\rho)_{45} P^{c,*}_{45,13}\rho_{32}\end{array}\right] + H.C.,$$

Similar to NEGF with Kadanoff–Baym equation (KBE) approximation at t=t'

Scattering Terms in Lindblad Dynamics

$$\frac{d\rho_{12}}{dt}\Big|_c = \frac{1}{2}\sum_{345}\left[\begin{array}{c}(I-\rho)_{13}\,P^{c}_{32,45}\rho_{45}\\ -(I-\rho)_{45}\,P^{c,*}_{45,13}\rho_{32}\end{array}\right] + H.C.$$

The sub-index "1", is a combined index of k-points and bands.

e-phonon

$$P^{\text{e-ph}}_{1234} = \sum_{q\lambda\pm} A^{q\lambda\pm}_{13} A^{q\lambda\pm,*}_{24}$$

$$A^{q\lambda\pm}_{13} = \sqrt{\frac{2\pi}{\hbar}}\, g^{q\lambda\pm}_{13} \sqrt{\delta^{G,\sigma}_{13}(\omega_{q\lambda})}\sqrt{n^{\pm}_{q\lambda}}$$

$g^{q\lambda}$ - e-ph matrix fully from first-principles with self-consistent SOC

e-impurity

$$P^{\text{e-i}}_{1234} = A^{i}_{13} A^{i,*}_{24}$$

$$A^{i}_{13} = \sqrt{\frac{2\pi}{\hbar}}\, g^{i}_{13} \sqrt{\delta^{G,\sigma}_{13}(0)}\sqrt{n_i V_{\text{cell}}}$$

$$g^{i}_{13} = \langle 1|\, V^{i}\, |3\rangle$$

1. DFT defect potentials V^i with supercell for neutral defect
2. Ionized impurity potential $V^{ion} = ZV^{scr}$ (+SOC corr.)

e-e

$$P^{\text{e-e}}_{12,34} = 2\sum_{56,78} (I-\rho)_{65}\, \mathscr{A}_{15,37}\mathscr{A}^{*}_{26,48}\rho_{78}$$

$$\mathscr{A}_{1234} = \frac{1}{2}\left(A_{1234} - A_{1243}\right)$$

$$A_{1234} = \sqrt{\frac{2\pi}{\hbar}}\, g^{e-e}_{1234} \sqrt{\delta^{G,\sigma}_{1234}}$$

$$g^{\text{e-e}}_{1234} = \langle 1|\langle 2|\, V(r,r')\, |3\rangle|4\rangle$$

Screened V (+SOC corr.) with RPA dielectric function

J. Xu, A. Habib, R. Sundararaman and Y. Ping, PRB, Editor's Suggestions, (2021).
J. Xu, A. Habib, S. Kumar, F. Wu, R. Sundararaman and Y. Ping, Nat. Commun. 11, 2780 (2020).

Lindbladian dynamics: R. Rosati et al, *Physical Review B*, **92**, 235423 (2015)

Verification of the Implementation of the Electron-Electron Scattering

- "FT-GW": the finite-temperature GW* method; compared to ImΣ_{ee} based on our used density-matrix dynamics

GW at finite T:

$$\langle n,\mathbf{k}|\mathrm{Im}\,\Sigma(\mathbf{r},\mathbf{r}';E)|n',\mathbf{k}\rangle$$

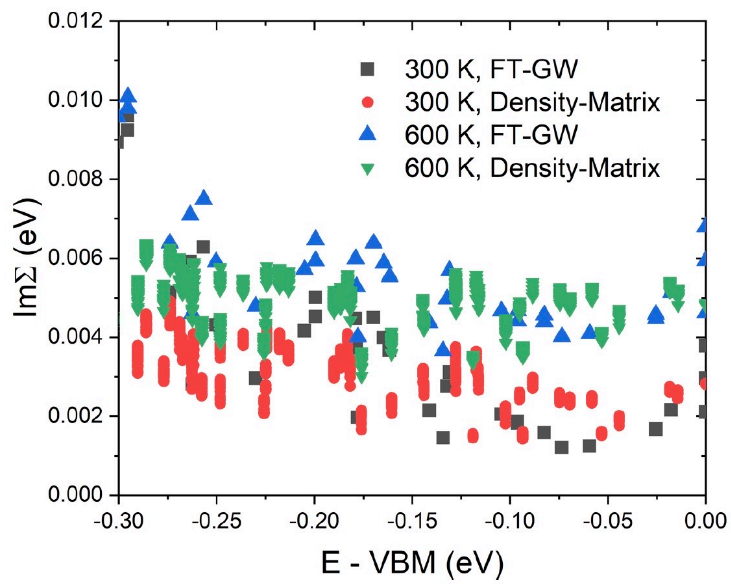
$$=\sum_{n_1}\sum_{\mathbf{q},\mathbf{G},\mathbf{G}'} M^{\mathbf{G}}_{n,n_1}(\mathbf{k},\mathbf{q})[M^{\mathbf{G}'}_{n',n_1}(\mathbf{k},\mathbf{q})]^* v(\mathbf{q}+\mathbf{G}') \times \mathrm{Im}\,\epsilon^{-1}_{\mathbf{G},\mathbf{G}'}(\mathbf{q},E-E_{n_1,\mathbf{k}-\mathbf{q}}) \times[1+n_B(E-E_{n_1,\mathbf{k}-\mathbf{q}})-n_F(E_{n_1,\mathbf{k}-\mathbf{q}})].$$

Density matrix dynamics at the semiclassical limit:

$$\frac{1}{\tau^c_{p,1}}=\sum_{2\neq 1}\left[P^c_{11,22}f_2+(1-f_2)\,P^c_{22,11}\right]$$

$$\mathrm{Im}\Sigma^c_1=\hbar/\left(2\tau^c_{p,1}\right)$$

*Lorin X. Benedict et al, PRB, 66, 085116 (2002)



- Two implementations agree well around the Fermi level

J. Xu…Y. Ping, Phys. Rev. B, (2021) Editor's Suggestions

Computational Scheme for Open Quantum Dynamics

DFT simulations
with spin-orbit coupling

Eigenstates, phonons,
e-phonon matrix with SOC
on coarse k and q meshes,
e.g., 24*24*24

JDFTx /
QuantumEspresso(EPW) → JDFTx

Wannier fitting
$|\psi_k\rangle \rightarrow |w_R\rangle$

Wannier interpolation
R space → k space

Eigenvalues, phonons,
e-phonon matrix
on fine k and q meshes,
e.g., 800*800*800

JDFTx

Density-matrix dynamics

$$\frac{d\rho}{dt} = \frac{d\rho}{dt}|_{\text{pump}} + \frac{d\rho}{dt}|_{\text{c}}$$

e-ph, e-e, e-i scattering matrix
Time evolution of observables
Spin, Carrier, Kerr rotation

$$\Delta S(t) = \Delta S(0)\exp\left(-\frac{t}{\tau_s}\right) \times\cos(\omega t)$$

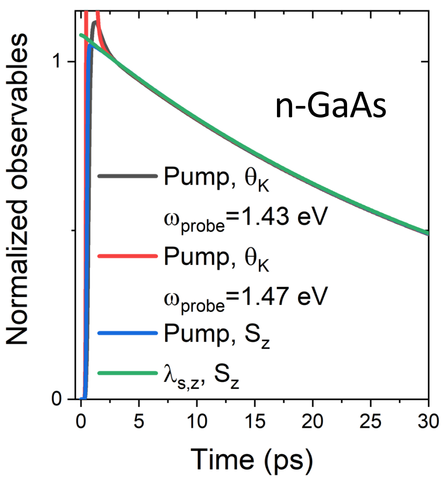
DMD code interfaced with JDFTx

$$\frac{d\rho_{12}}{dt}|_c = \frac{1}{2}\sum_{345}\left[\begin{array}{c}(I-\rho)_{13}\,P^{c}_{32,45}\rho_{45}\\ -(I-\rho)_{45}\,P^{c,*}_{45,13}\rho_{32}\end{array}\right] + H.C.$$

J. Xu, A. Habib, R. Sundararaman and Y. Ping, PRB, Editor's Suggestions, (2021).
J. Xu, A. Habib, S. Kumar, F. Wu, R. Sundararaman and Y. Ping, Nat. Commun. 11, 2780 (2020).

Physical Observables

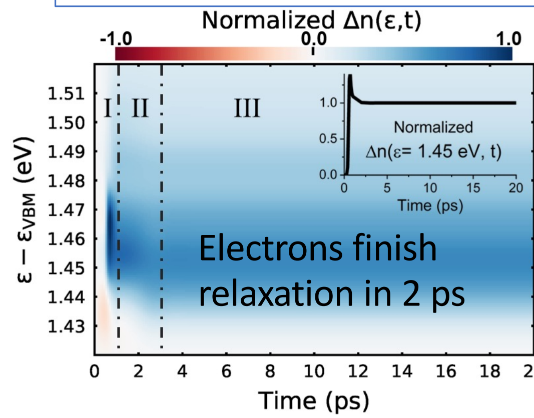
Time resolved Kerr rotation

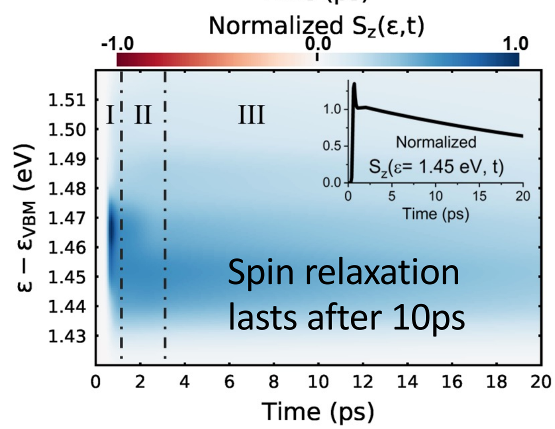


Calculated $\tau_s = 13$ps
at RT vs exp. spin lifetime ~10ps

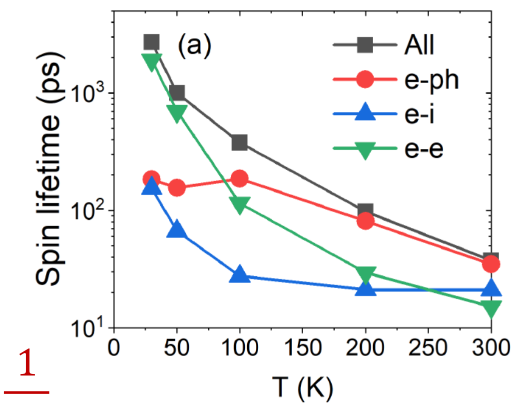
$$\theta_K = \mathrm{Im}\frac{\sqrt{\epsilon_+} - \sqrt{\epsilon_-}}{1 - \sqrt{\epsilon_+}\sqrt{\epsilon_-}}$$

Coupled electron and spin relaxation

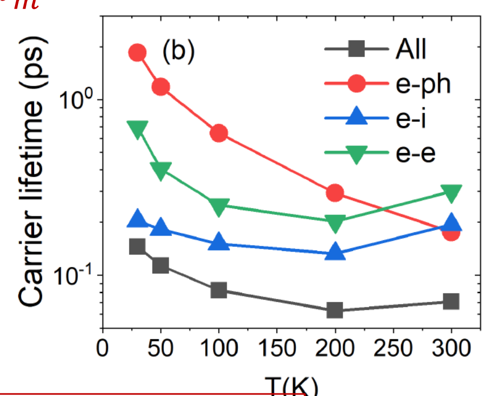




Mechanistic study



$$\tau_s \propto \frac{1}{\tau_m}$$

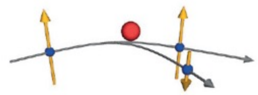


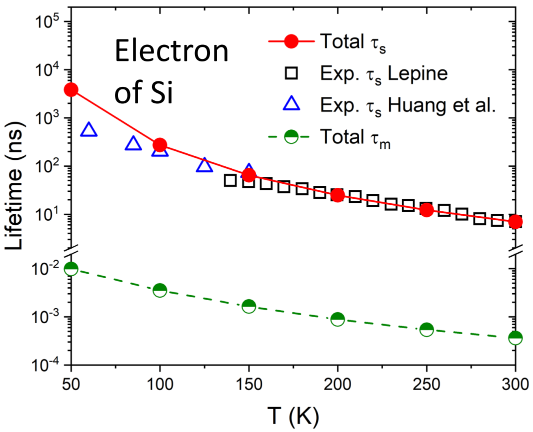
J. Xu...Y. Ping, PRB, **104**, 184418, (2021), Editors' suggestions.

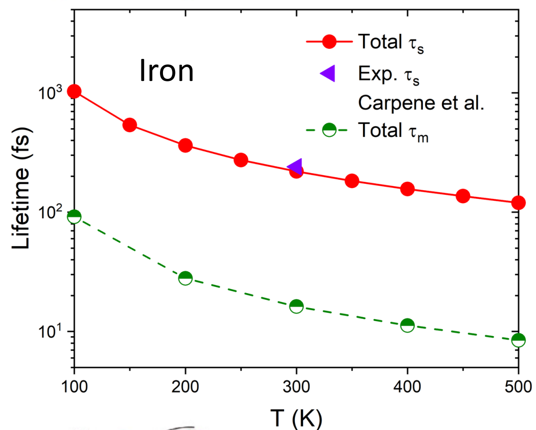
Study dependence on finite E and B fields, T, and carrier density

Spin Dynamics for General Solids

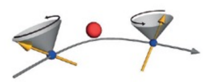
With Inversion Symmetry

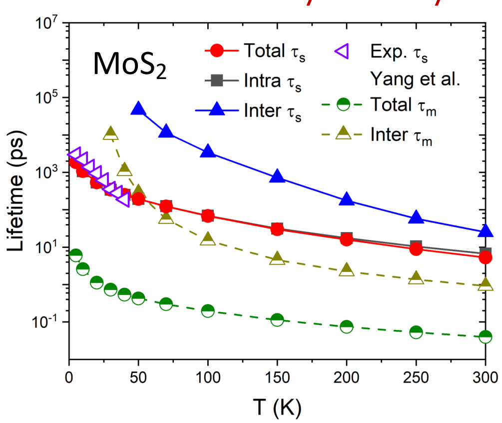


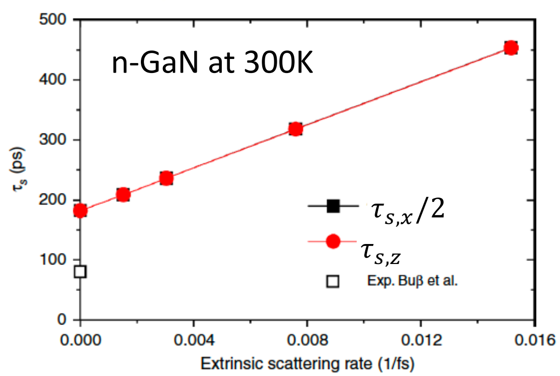




No Inversion Symmetry



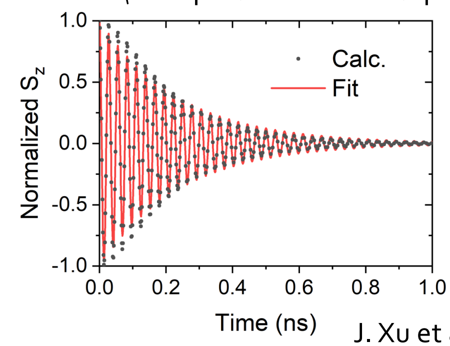


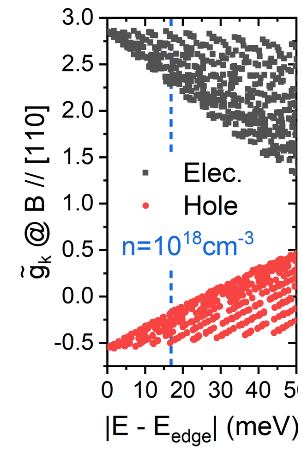


Ensemble spin dephasing T_2* in $CsPbBr_3$ at 4K, B=0.5T

$$H_k = H_{0,k} + \mu_B \mathbf{B} \cdot (\mathbf{L}_k + g_0 \mathbf{S}_k)$$

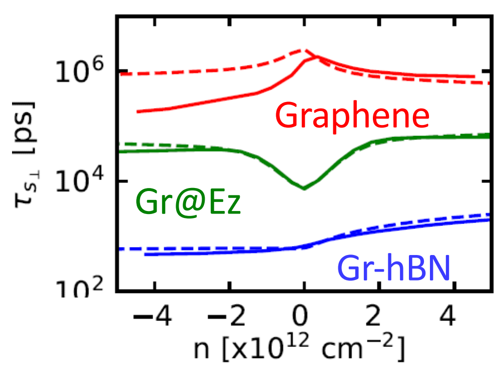
$$L_{kmn} = i \left\langle \frac{\partial u_m}{\partial \mathbf{k}} \right| \times \left(\hat{H} - \frac{\varepsilon_m + \varepsilon_n}{2} \right) \left| \frac{\partial u_n}{\partial \mathbf{k}} \right\rangle$$

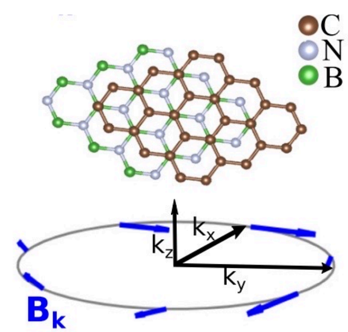




J. Xu et al (2022), arXiv:2210.17074

Graphene spin relaxation





J. Xu… Y. Ping, Phys. Rev. B, (2021) Editor's Suggestions; Nat. Commun. (2020); Nano Letters (2021); A. Habib et al, Phys. Rev. B (2022)

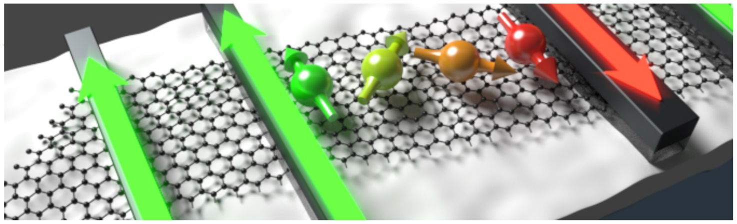
Outline

- Introduction of spin-based QIS
- Theoretical framework for ab-initio spin dynamics in solids
- **Spin relaxation and transport in 2D materials**
- Spin relaxation and dephasing in halide perovskites
- Conclusion and outlook

Design Materials for Optimal Spin Transport

Graphene

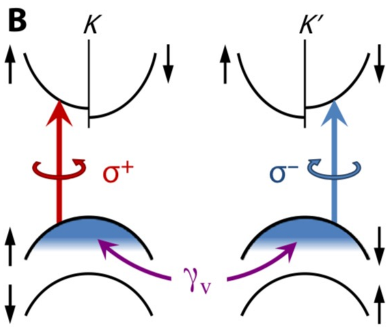
- ✓ High carrier mobility
- ✓ Longest RT spin diffusion length
- ✗ No spin-valley locking (too small SOC)



https://institut2a.physik.rwth-aachen.de/research

Transition metal dichalcogenides

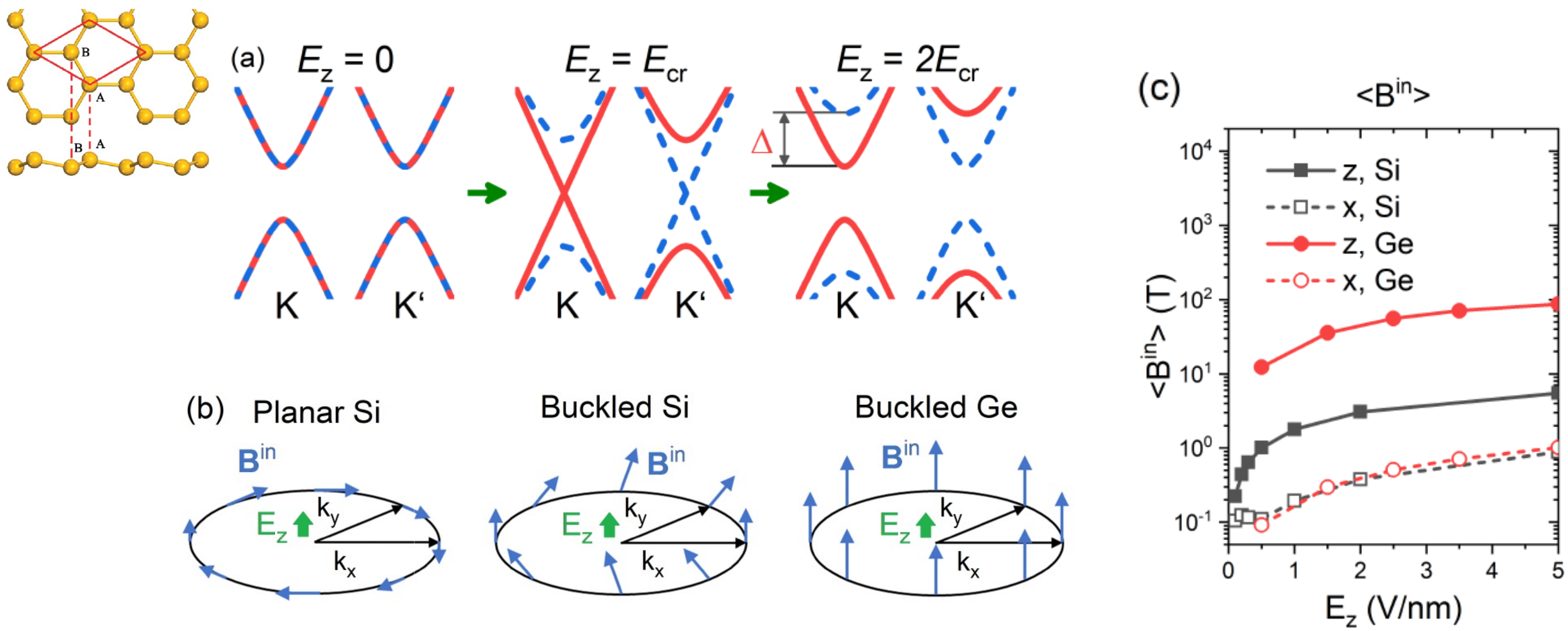
- ✓ Spin-valley locking effect
- ✓ Ultralong spin lifetime at low T
- ✗ Low carrier mobility



Goryca et al., Sci. Adv., 5, eaau4899 (2019)

Can we have a material with spin-valley locking,
high carrier mobility, long spin lifetime and spin diffusion length?

Spin-orbit Coupling in Silicene and Germanene under E field

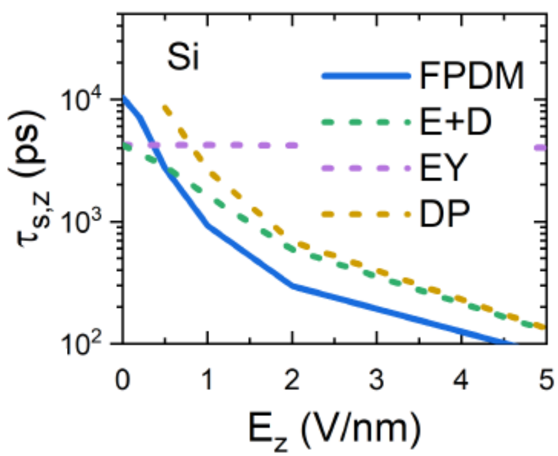


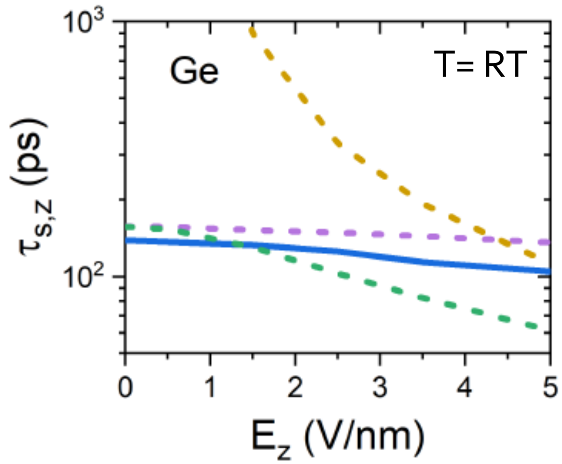
- Spin is strongly polarized along z in bulked germanene under E_z field

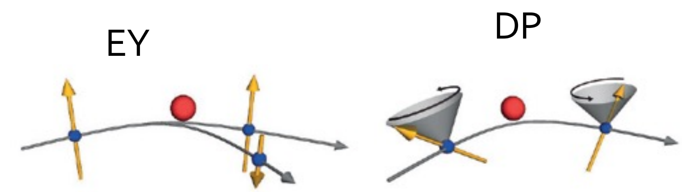
$$\mathrm{B}_{kn}^{\mathrm{in}} = 2\Delta_{kn}\mathrm{S}_{kn}^{\mathrm{exp}} / (g_e\mu_B)$$

J. Xu, A. Habib, R. Sundararaman, Y. Ping, Nano Letters, **21**, 9594, (2021)

Spin-orbit Coupling in Silicene and Germanene under E field





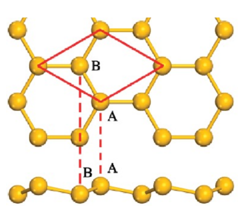


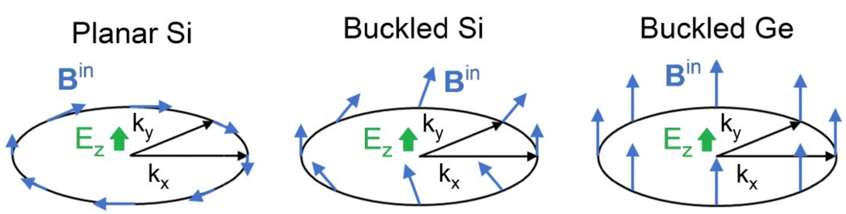
$$\left(\tau_{s,i}^{\mathrm{EY}}\right)^{-1} \approx 4\left\langle b_i^2\right\rangle\left\langle\tau_p^{-1}\right\rangle$$

$$\left(\tau_{s,i}^{\mathrm{DP}}\right)^{-1} \approx \left\langle\tau_p^{-1}\right\rangle^{-1}\left\langle\Omega^2-\Omega_i^2\right\rangle$$

$$\Omega_i = g_e \mu_B B_i^{\mathrm{in}}$$

- Silicene under E is closer to DP
- Germanene under E is closer to EY

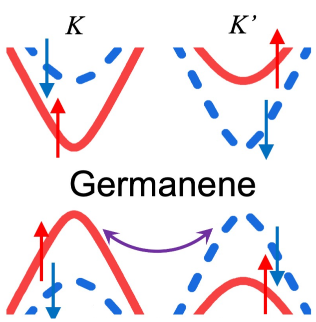




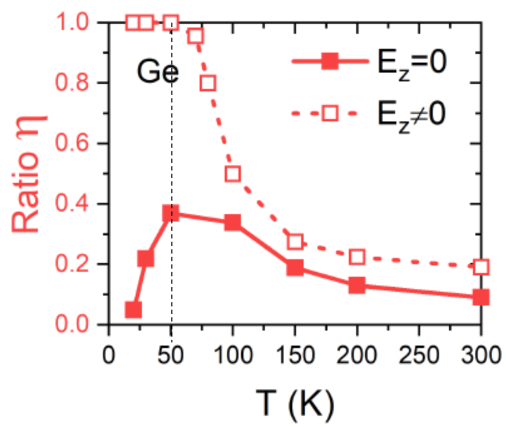
J. Xu et al, Nano Letters (2021), arXiv: 2110.01128

Spin-Valley Locking and Spin Transport of Germanene under E

Spin-valley locking



intervalley contribution ratio



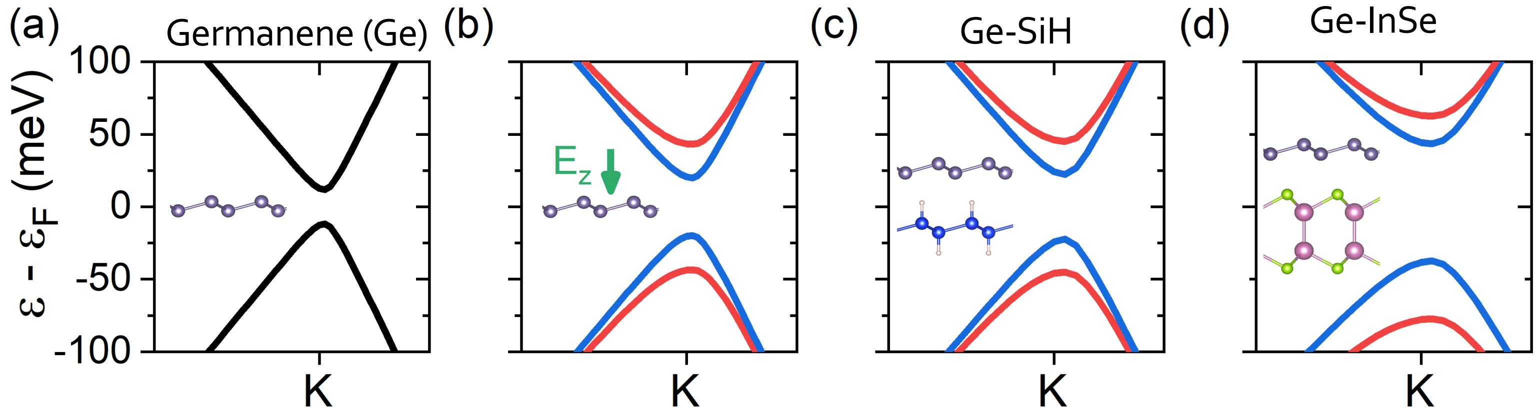
Ab-initio spin and carrier transport calculations under E field

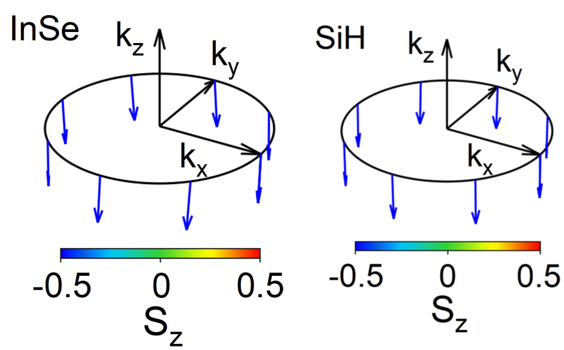
T (K)	n_i (cm^{-2})	τ_p (ps)	$\overline{\mu_c}$ (cm^2/(V s))	D (cm^2/s)	$\tau_{s,z}$ (ns)	$l_{\parallel,s_z}$ (μm)
300	0	0.4	3.2×10^4	830	0.1	2.9
300	1×10^{11}	0.3	2.5×10^4	620	0.1	2.5
50	0	7.2	3.8×10^6	16700	97	400
50	1×10^{11}	1.6	4.5×10^5	2000	76	120
50	1×10^{12}	0.2	5.8×10^4	250	26	25

- Spin valley locking: spin and valley polarization changes together; intervalley spin flip transition dominates
- Long spin lifetime 100 ns at low impurity density due to spin-valley locking
- Long spin diffusion length $l_{||,Sz} = \sqrt{D.\tau_{S,Z}} \approx 120\ \mu m$ at moderate impurity density $10^{11} cm^{-2}$ vs. graphene 1-40 μm

J. Xu, A. Habib, R. Sundararaman, Y. Ping, Nano Letters, **21**, 9594, (2021)

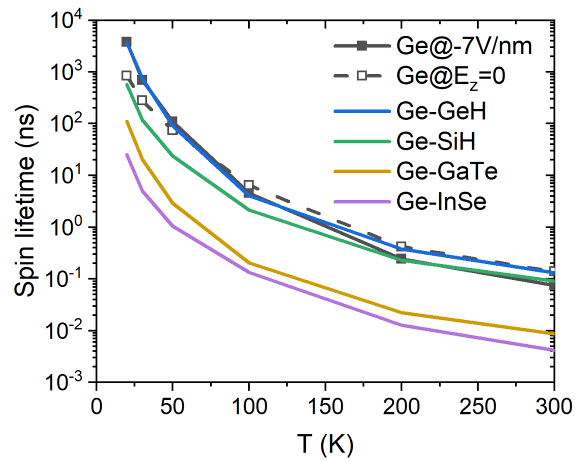
Substrate Effect on Spin Relaxation of Strong SOC Systems





tilted spin texture

vertical spin texture

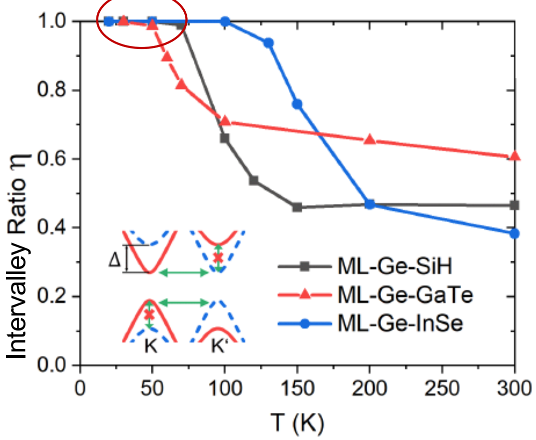


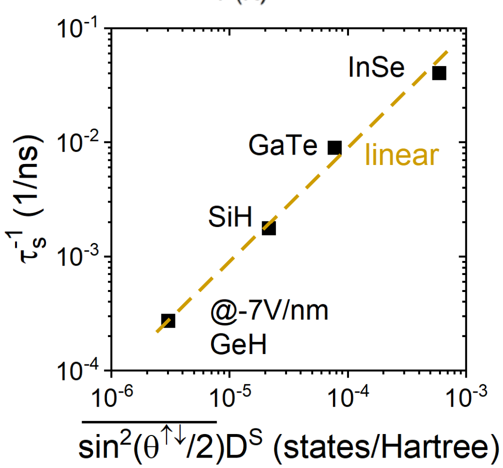
How different substrates affect spin relaxation differently?

J. Xu, Y. Ping, npj Comput. Mater. in press (2023), arXiv:2206.00784

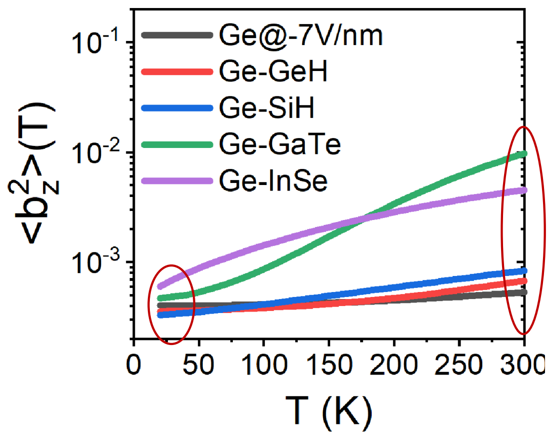
Substrate Effect on Spin Relaxation of Strong SOC Systems

Spin-valley locking below 20K



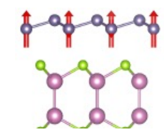


Spin mixing only explains high temperature spin lifetime difference



$$\left(\tau_{s,i}^{\mathrm{EY}}\right)^{-1} \approx 4\left\langle b_i^2\right\rangle\left\langle\tau_p^{-1}\right\rangle$$

Dominate phonon at K nearly unchanged



Substrate	ω_K (meV)	Contribution
Ge@-7V/nm	7.7	78%
Ge-GeH	6.9	70%
Ge-SiH	7.1	64%
Ge-GaTe	6.4	90%
Ge-InSe	7.2	99%

$\theta_{k_1,k_2}^{\uparrow\downarrow}$: angle between pair of spin states (k_1, k_2) spin expectation value direction

Spin flip angle $\theta_{k_1,k_2}^{\uparrow\downarrow}$ has best correlation with spin lifetime as a function of substrates

J. Xu, Y. Ping, npj Comput. Mater. in press (2023), arXiv:2206.00784

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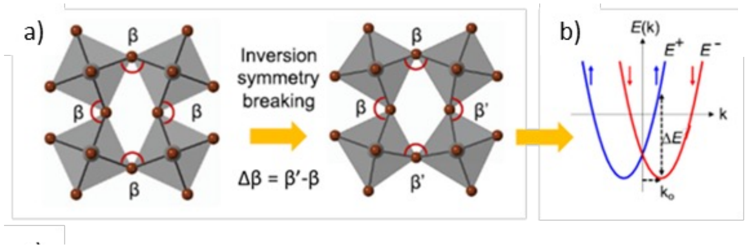
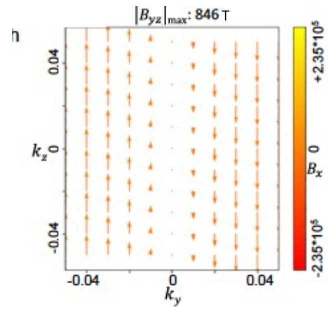
Opto-Spintronics in Hybrid Organic-Inorganic Materials

Rashba splitting tuning

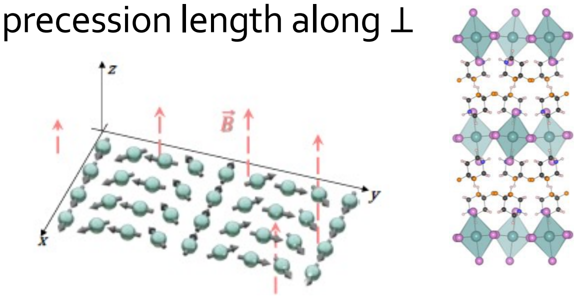


Manoj Jana et al, Nat. Commun. 2021

Persistent spin helix in 2D hybrid perovskite

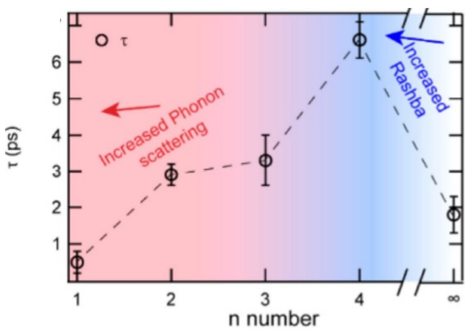


Maximum spin diffusion length in || direction >> spin precession length along ⊥



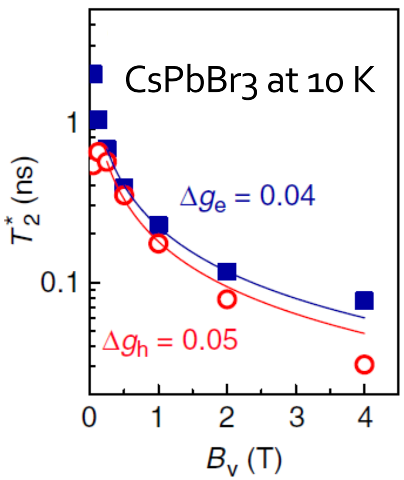
Lifu Zhang et al, Nat. Photonics, 2022

Spin relaxation in 2D perovskite



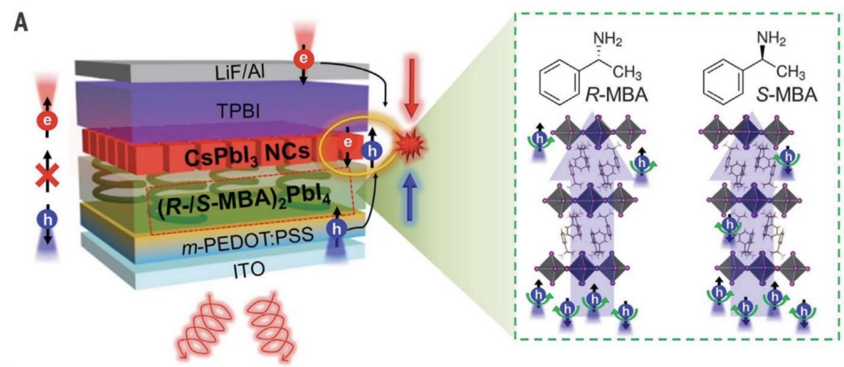
Exp. τ_s with thickness

X. Chen et al, ACS Energy Lett. 2018.



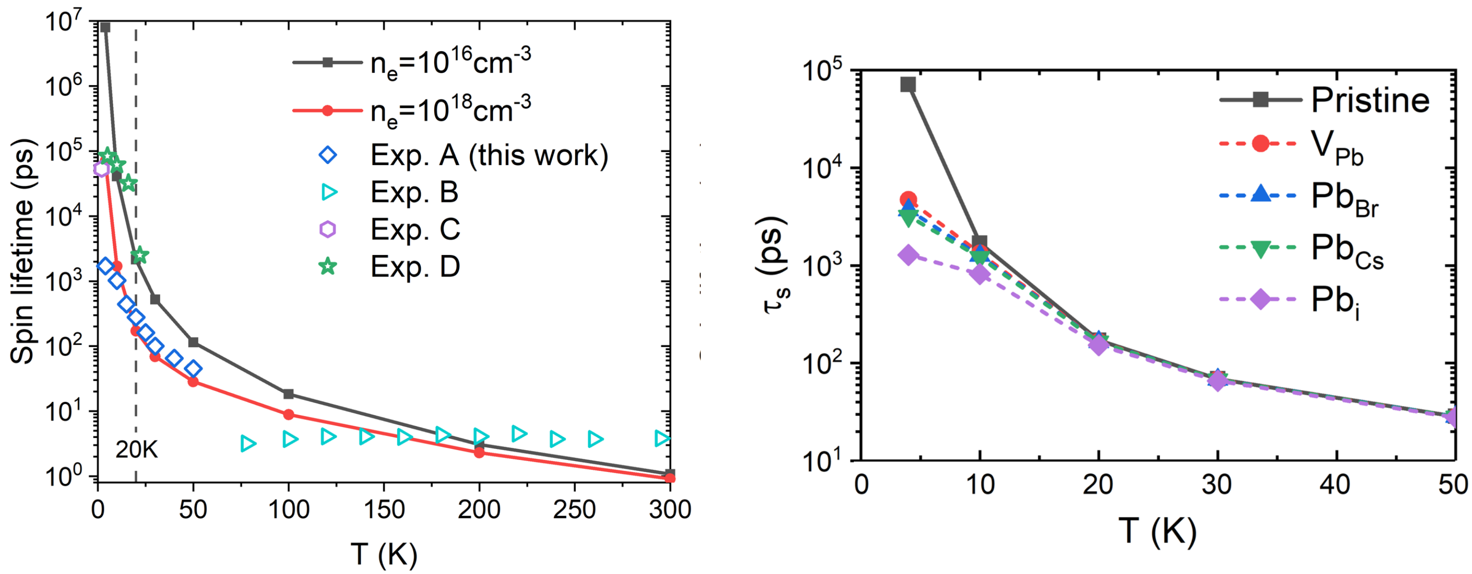
V. Belykh et al, Nat. Commu. 2019.

Chiral induced spin selectivity



Kim et al., Science 371, 1129–1133, (2021)

Spin Relaxation and Decoherence of CsPbBr3

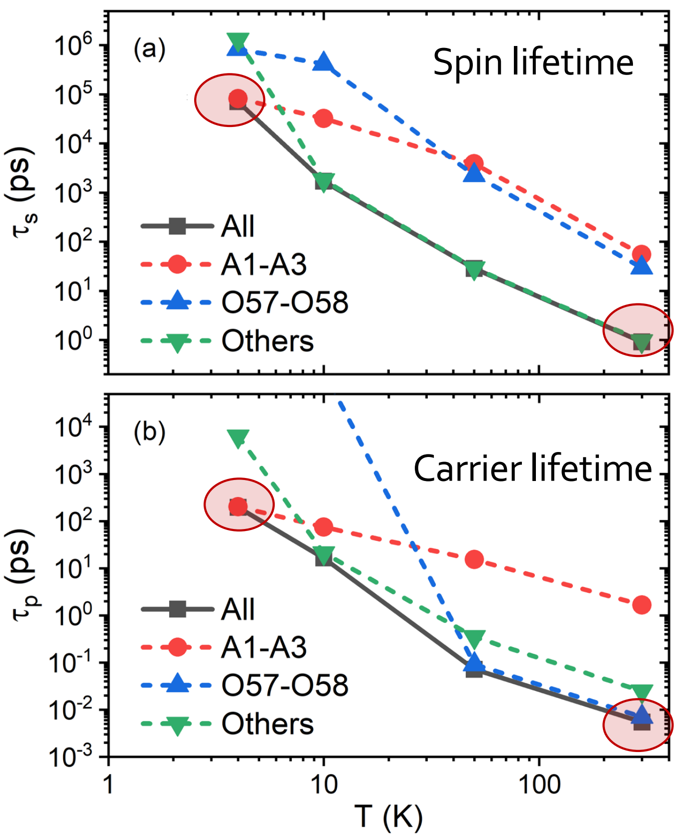


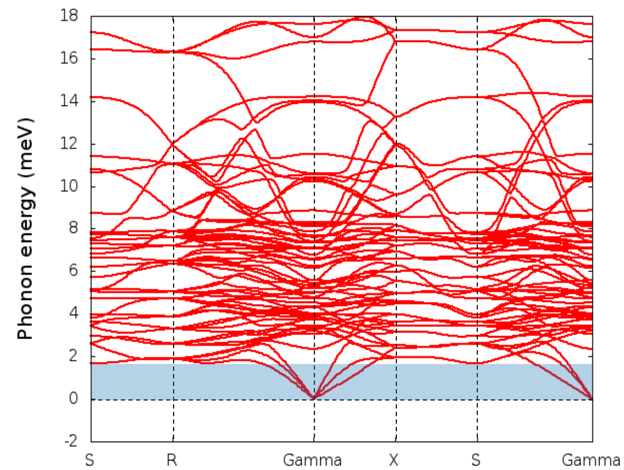
- Good agreement between theory and experiments > 20 K
- We found spin lifetime has strong dependence on carrier density (exp: n ~10^{18} cm^{-3}) <20 K
- Below 20K, defects/impurities have important contributions

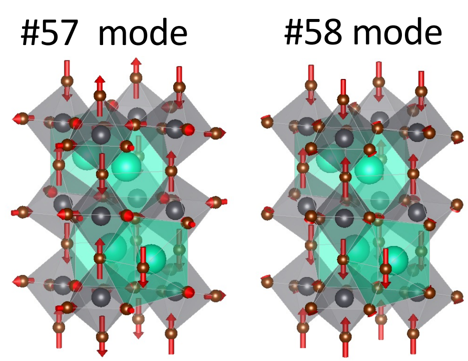
J. Xu, K. Li, U. Huynh, J. Huang, R. Sundararaman, V. Vardeny, Y. Ping, *Nat. Commun.* under review (2022)

Exp.A: experiment in this work
Exp B: Zhou et al., J. Phys. Chem. Lett. 11, 1502, (2020)

Different Phonon Dependence for Spin and Carrier lifetime







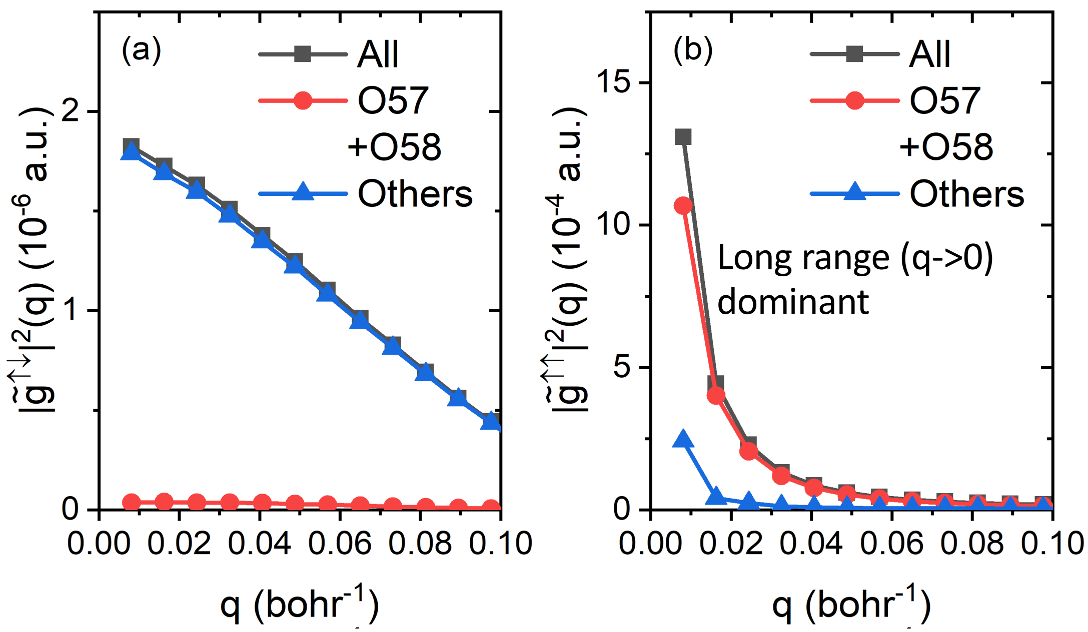
- Acoustic phonon (A1-A3) dominates at low temperature
- Optical modes (O57-O58; mixed LO-TO) dominant in carrier relaxation, but not in spin relaxation

J. Xu, K. Li, U. Huynh, J. Huang, R. Sundararaman, V. Vardeny, Y. Ping, *Nat. Commun.* under review (2022)

Fröhlich Electron-Phonon Coupling for Spin and Carrier lifetime

Related to spin relaxation

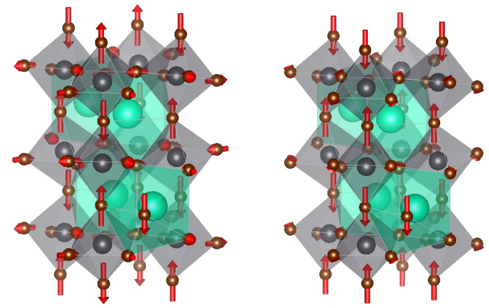
Related to carrier relaxation



Fröhlich e-ph is long ranged and spin independent -

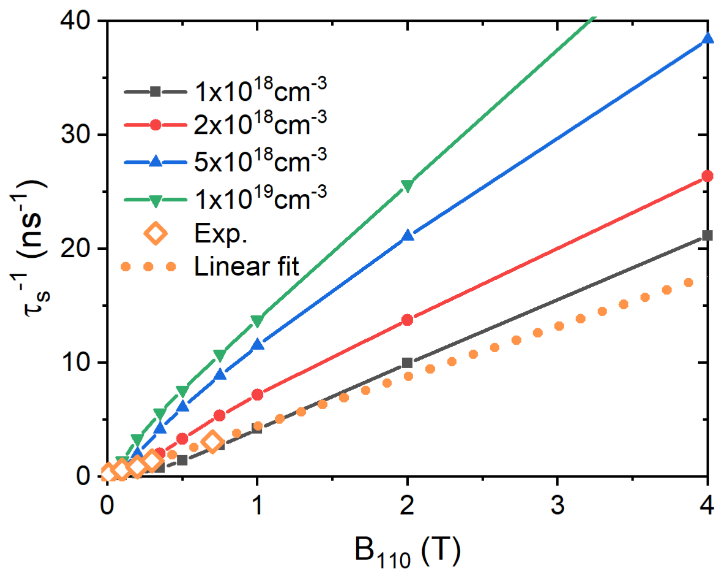
dominates carrier relaxation, NOT spin relaxation

#57 mode #58 mode



J. Xu, K. Li, U. Huynh, J. Huang, R. Sundararaman, V. Vardeny, Y. Ping, *Nat. Commun.* under review (2022)

Ensemble Spin Dephasing T2* under B Field of CsPbBr3



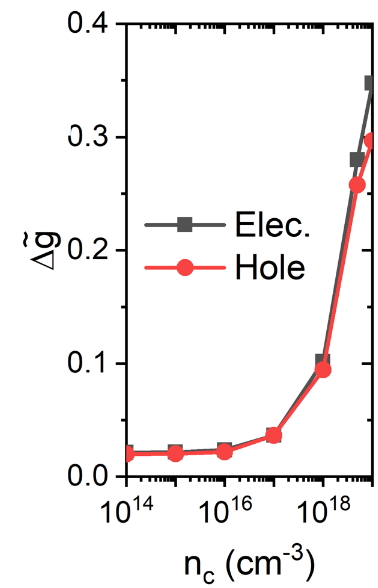
(T=20K, exp. carrier density ~10^{18} cm^{-3})

- At relatively high B field, spin relaxation rate linearly proportional to B field

$$\tau_s^{-1}(\mathbf{B}) \approx (\tau_s^0)^{-1} + (\tau_s^{\Delta\Omega})^{-1}(\mathbf{B})$$

$$(\tau_s^{\Delta\Omega})^{-1} \sim (\tau_s^{\mathrm{FID}})^{-1} \sim C^{\Delta g}\Delta\Omega = C^{\Delta g}\mu_B B\Delta\tilde{g},$$

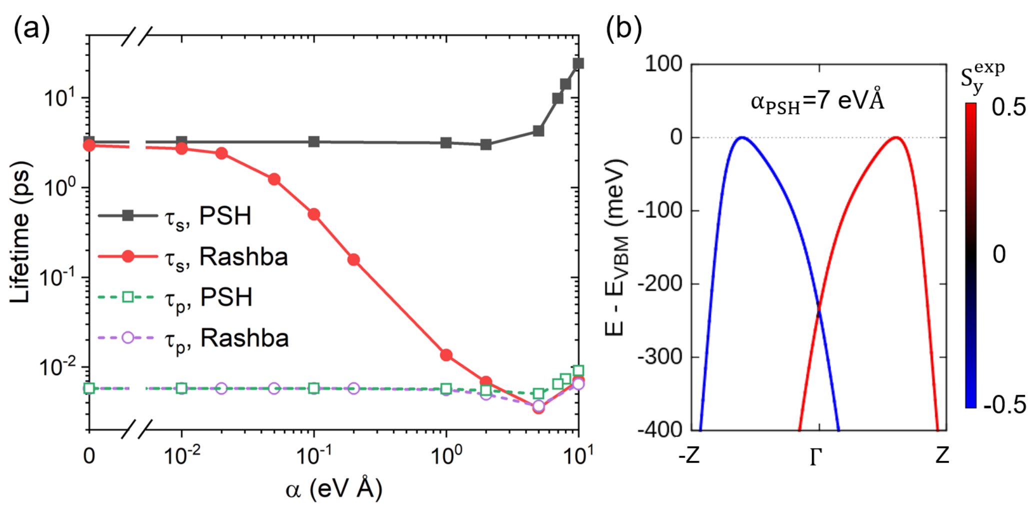
- The strength of B field dependence varies with carrier density due to $\Delta\tilde{g}$

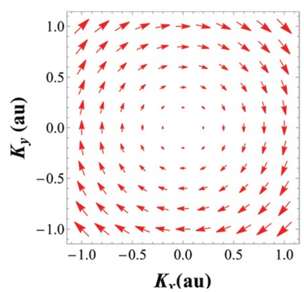


Efficient tuning spin T_2*
by changing carrier density

J. Xu, K. Li, U. Huynh, J. Huang, R. Sundararaman, V. Vardeny, Y. Ping, *Nat. Commun.* under review (2022)

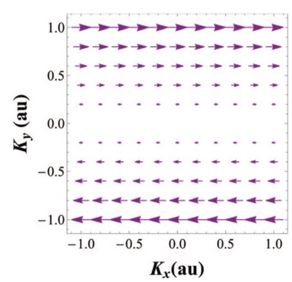
Relation of Spin Texture and Spin Lifetime





Rashba

$\vec{\Omega}_k = \alpha^R(\sigma\times\mathbf{p}).\hat{z}$



Persistent Spin Helix

$\vec{\Omega}_k = \alpha^{PSH}k_y\hat{z}$

- Including broken-inversion symmetry SOC to $CsPbBr_3$ perturbatively for extrinsic effects
- PSH spin texture can increase spin lifetime when SOC strength is large, not Rashba
- PSH spin texture can be realized by controlling crystal symmetry*

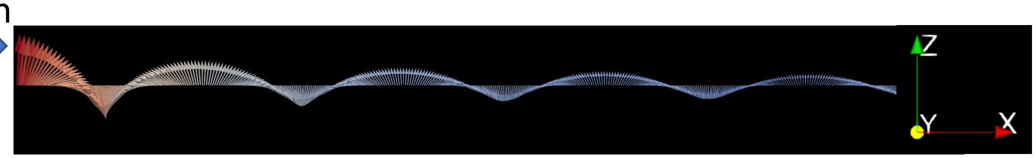
*Lifu Zhang et al, Nat. Photonics, 2022

J. Xu, K. Li, U. Huynh, J. Huang, R. Sundararaman, V. Vardeny, Y. Ping, *Nat. Commun.* under review (2022)

Work in Progress

Spatial-resolved spin transport:

Spin injection



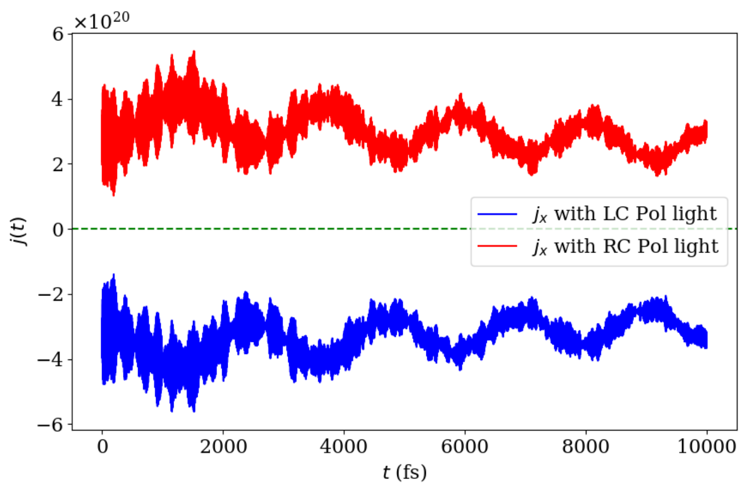
$$\frac{\partial \rho_{\mathbf{k},nm}(\mathbf{R},t)}{\partial t} + \frac{1}{2}\left\{\frac{\partial H_0}{\partial \mathbf{k}}, \frac{\partial \rho_{\mathbf{k}}(\mathbf{R},t)}{\partial \mathbf{R}}\right\}_{nm}$$

$$= \frac{1}{2}\left\{\dot{\mathbf{k}}, \frac{\partial \rho_{\mathbf{k}}(\mathbf{R},t)}{\partial \mathbf{k}}\right\}_{nm} + \frac{\partial \rho_{\mathbf{k},nm}(\mathbf{R},t)}{\partial t}\Big|_{\mathrm{light}} + \frac{\partial \rho_{\mathbf{k},nm}(\mathbf{R},t)}{\partial t}\Big|_{\mathrm{scatt}},$$

(understanding CISS effect)

Predict spin coherence and diffusion length, and time-dependent spin texture and polarization

Steady-state photocurrent (photogalvanic effect):



$$j_x = Tr(\rho v_x)$$

Density matrix dynamics with **absorption, scattering, stimulated and spontaneous emission** for photocurrents (shift current and CPGE) in noncentral-symmetric materials

Conclusion

- Developed density matrix formalism for open systems including electron-phonon, e-e, e-i interactions and spin-orbit for coupled spin and carrier dynamics in general solids

J. Xu, A. Habib, S. Kumar, R. Sundararaman, Y. Ping, *Nat. Commun*. **11**, 2780, (2020).
J. Xu, A. Habib, R. Sundararaman, Y. Ping, *PRB*, **104**, 184418, (2021), *Editor's suggestions*

- Realized spin-valley locking and long spin lifetime in 2D Dirac materials under E field and investigated substrate effects, and investigated g factor fluctuation effect on spin dephasing

J. Xu, H. Takenaka, A. Habib, R. Sundararaman, Y. Ping, *Nano Letters,* **21**, 9594, (2021)
J. Xu, Y. Ping, *npj Comput. Mater*. in press, 2023, arXiv:2206.00784
J. Xu, K. Li, U. Huynh, J. Huang, V. Vardeny, R. Sundararaman, Y. Ping, *Nat. Commun.* under review (2022) https://arxiv.org/abs/2210.17074

- Work in progress on developing methods for computing steady-state photocurrents for photogalvanic effect, and developing spatial resolved spin transport to understand the effect of CISS

Postdoc and PhD positions are available!

NSF DMR-1956015

Computational codes

- DFT calculations
- BSE code built on a private version of Quantum Espresso
- GW implementation for 2D built on the West code; benchmarked with the Yambo code
- Density matrix and spin dynamics in DMD code interfaced with JDFTx code and EPW