Нелинейный классический и линейный квантовый транспорт экситонов в двумерных кристаллах

М.М. Глазов

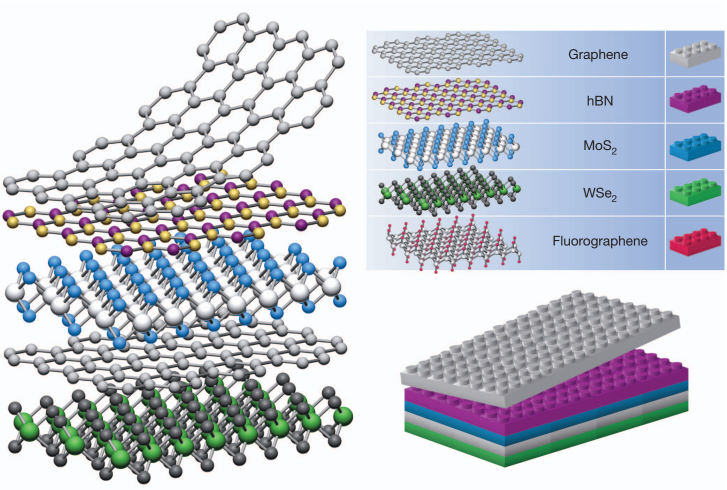
ФТИ им. А.Ф. Иоффе, Санкт-Петербург

1. Краткое введение в физику двумерных кристаллов
2. Нелинейный транспорт экситонов
3. Слабая локализация экситонов
4. Заключение

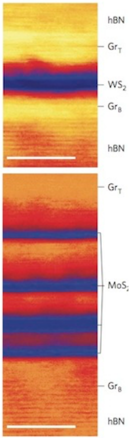
Нелинейные волны-2020, 5 марта 2020. Лекция №39

Two-dimensional materials

are the platform for van der Waals heterostructures

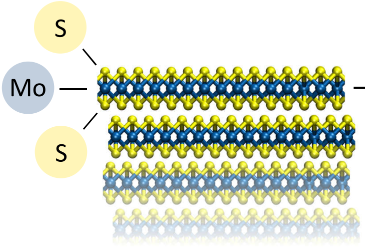


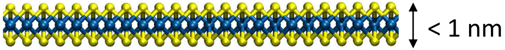
Geim, Grigorieva (2013)



Withers et al. (2015)

Transition metal dichalcogenides



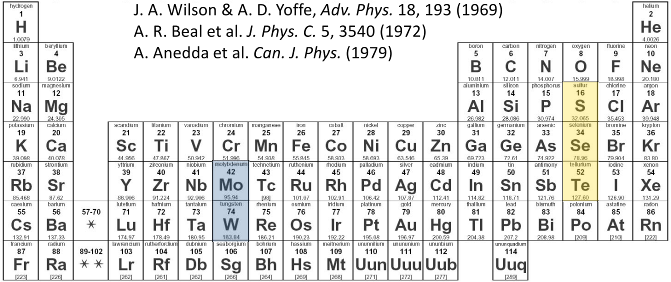


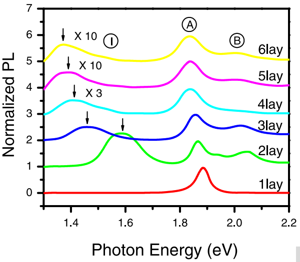
Monolayers are **direct**-gap semiconductors

A. Splendiani et al., *Nano Lett.* 10, 1271 (2010)
K. F. Mak et al., *Phys. Rev. Lett.* 105, 136805 (2010)

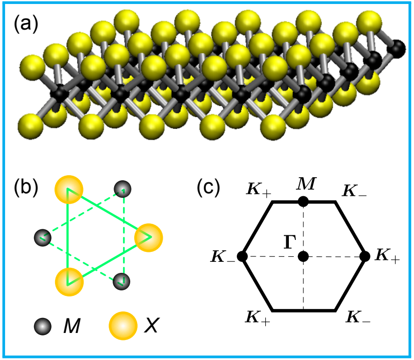
Bulk materials are **indirect**-gap semiconductors

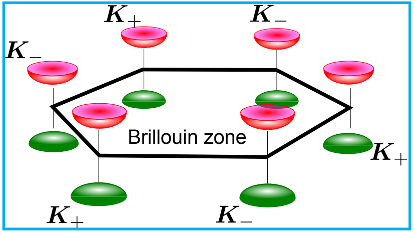
B. L. Evans & P. A. Young, *Proc. Phys. Soc.* 91, 475 (1967)
J. A. Wilson & A. D. Yoffe, *Adv. Phys.* 18, 193 (1969)
A. R. Beal et al. *J. Phys. C.* 5, 3540 (1972)
A. Anedda et al. *Can. J. Phys.* (1979)





Direct and inverse lattices of MX_2





MoS_2, $MoSe_2$, WS_2, WSe_2, …

D_{3h} point symmetry: horizontal reflection plane (σ_h), three-fold rotation axes (C_3, S_3), $3C_2$, $2\sigma_v$

no space inversion: SO-splitting, second harmonic generation

Hexagonal Brillouin zone, direct band gaps are formed at the $\boldsymbol{K}_+$ and $\boldsymbol{K}_-$ edges

Valley symmetry is C_{3h}; valleys are chiral

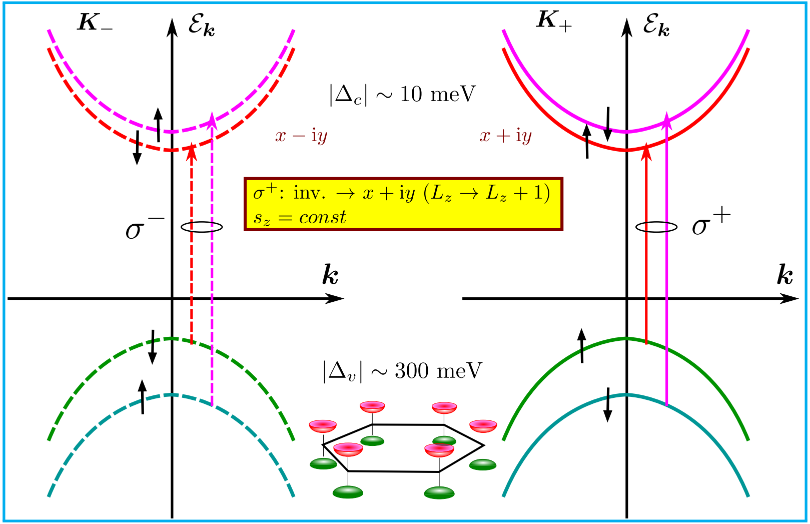
$\boldsymbol{K}_+$ and $\boldsymbol{K}_-$ valleys are related by the time reversal symmetry

$$\mathcal{H}_{\text{eff}} = \hbar v(\boldsymbol{\sigma} \cdot \boldsymbol{k}) + \frac{E_g}{2}\sigma_z$$

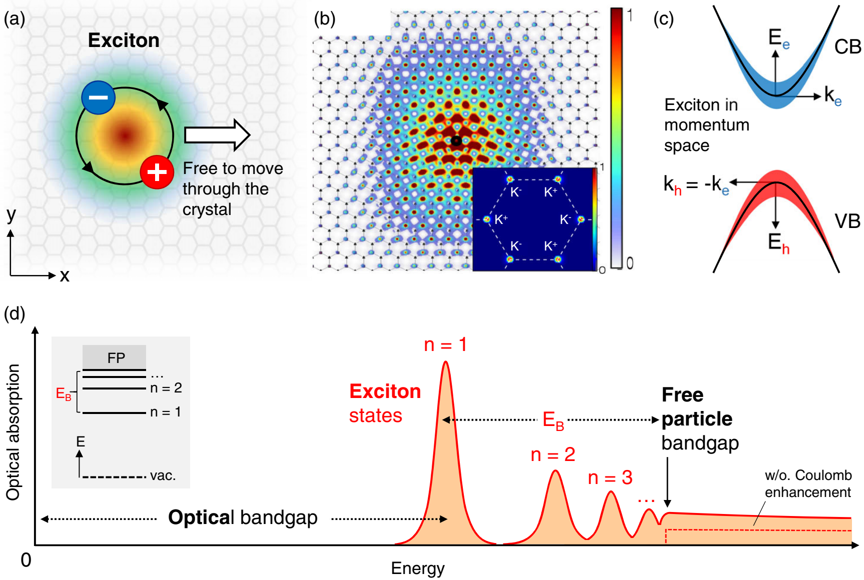
Review: Rev. Mod. Phys. **90**, 021001 (2018); Phys. Usp. **61**, 825 (2018)

Band structure & selection rules

Mo-based



Excitons in atom-thin semiconductors



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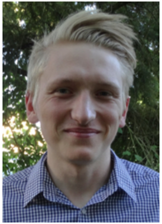
Эксперимент: Университет Регенсбурга, Германия

Alexey Chernikov

Marvin Kulig

Jonas Zipfel

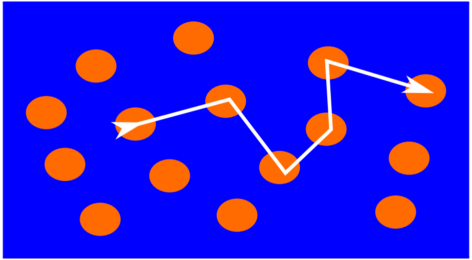
Jonas Ziegler



Rudolf Richter

Transport & optical effects

Velocity: $\delta v(t) = \delta v(0) e^{-t/\tau_p}$

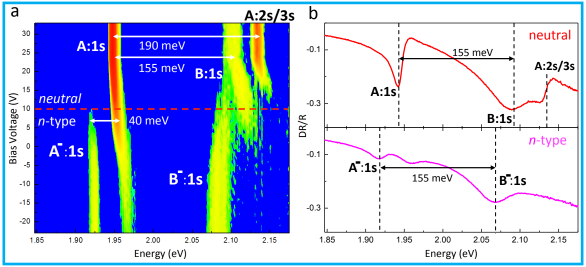


Velocity fluctuations:

$$\langle \delta v_x(t) \delta v_x(0) \rangle = \frac{v_T^2}{2} e^{-|t|/\tau_p} \quad (2D)$$

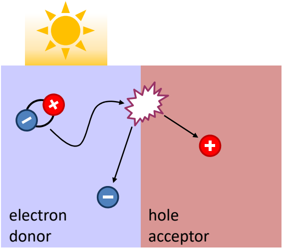
Diffusion coefficient

$$D = \int_0^\infty \langle \delta v_x(t) \delta v_x(0) \rangle dt = \frac{v_T^2 \tau_p}{2}$$

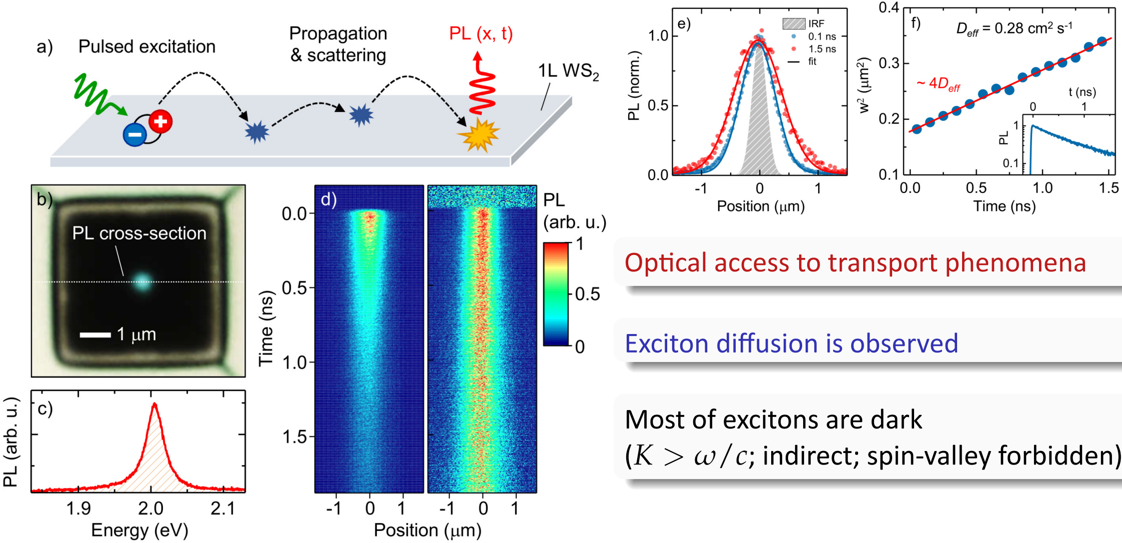


Phys. Rev. Materials **2**, 011001(R) (2018)

Prospects: solar energy harvesting



Exciton diffusion: experimental puzzles



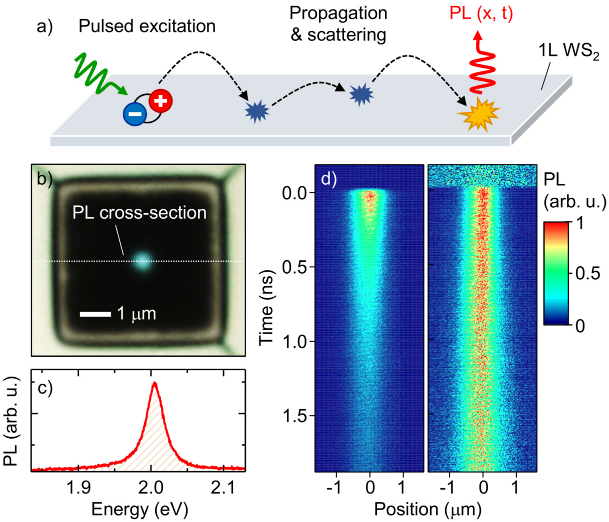
Optical access to transport phenomena

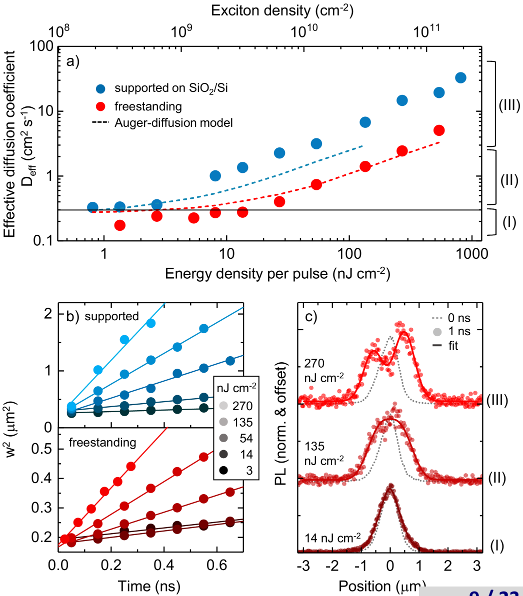
Exciton diffusion is observed

Most of excitons are dark
($K > \omega / c$; indirect; spin-valley forbidden)

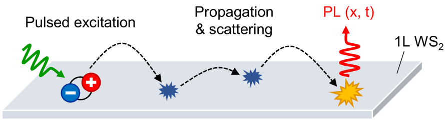
Phys. Rev. Lett. **120**, 207401 (2018)

Exciton diffusion: experimental puzzles

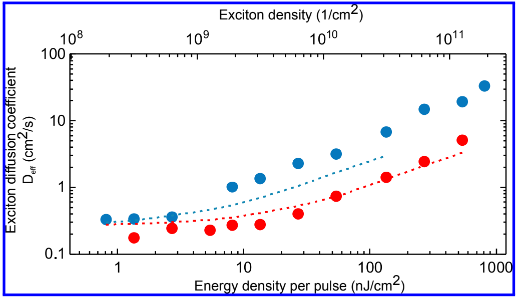




Phys. Rev. Lett. **120**, 207401 (2018)



Experiment & Numerics:



Diffusion equation

$$\frac{\partial n}{\partial t} = D\Delta n - \frac{n}{\tau} - R_A n^2 \quad R_A = (0.1\ldots0.5)\,\frac{\mathrm{cm}^2}{\mathrm{s}}$$

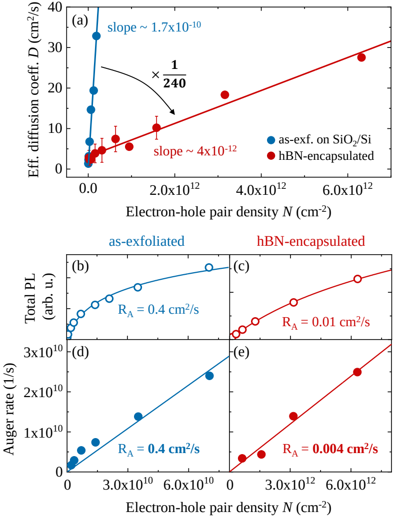
Ansatz solution

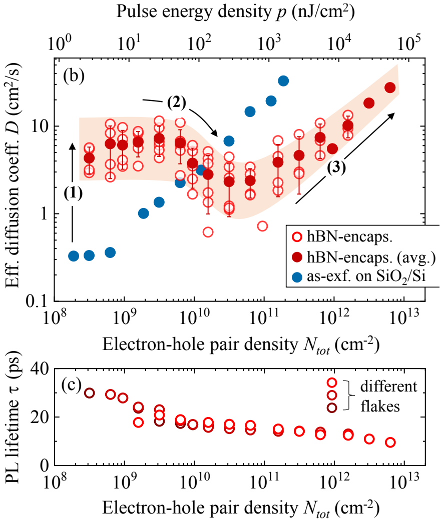
$$n(\rho,t) = \frac{N(t)}{\pi(r_0^2 + 4D_{eff}t)}\exp\left(-\frac{\rho^2}{r_0^2+4D_{eff}t}\right), \quad D_{eff} = D + \frac{R_A n_0 r_0^2}{16}$$

Encapsulation effect

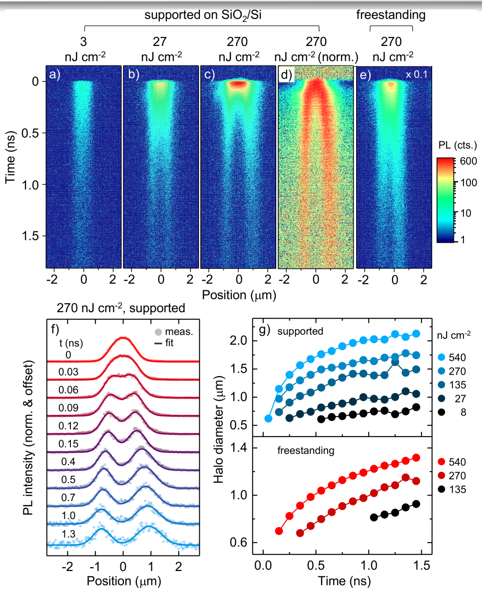
Suppression of Auger recombination effect $\Rightarrow$ weaker increase of $D_{eff}(N)$

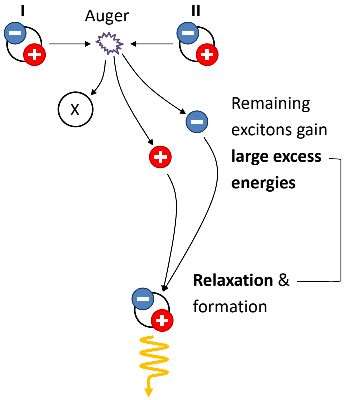
arXiv:1911.02909 (2019)





Excitonic halos: memory and heating effects

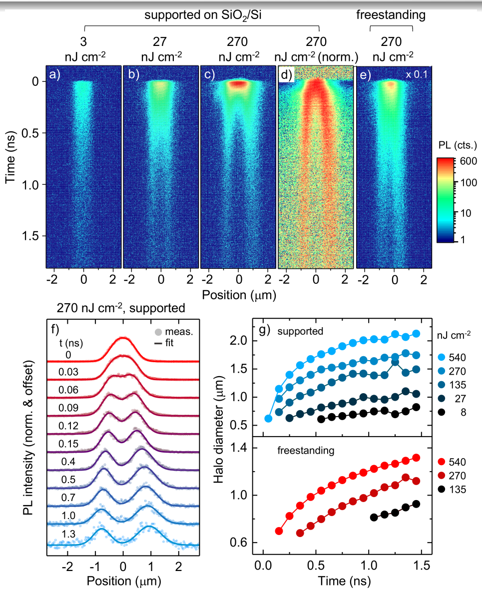


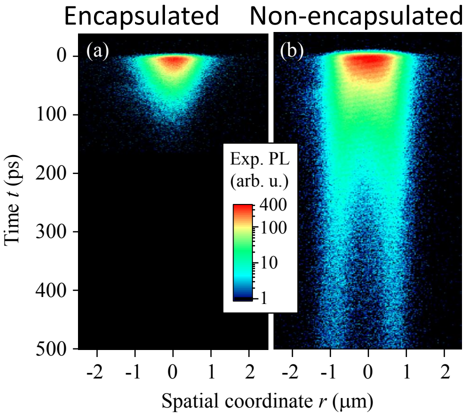


Excitons are overheated as a result of the Auger process non-equilibrium phonons are produced

Phys. Rev. Lett. **120**, 207401 (2018)

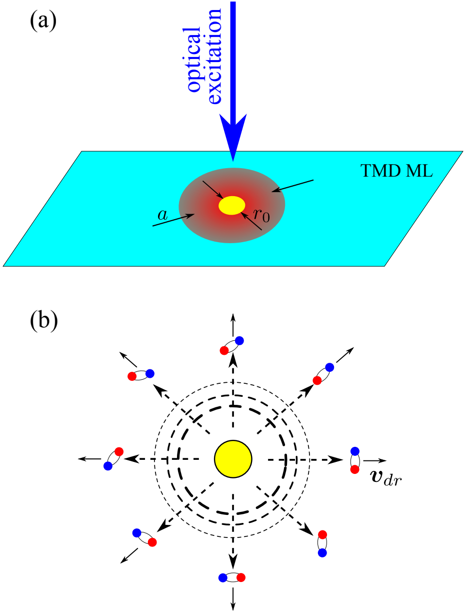
Excitonic halos: memory and heating effects





arXiv:1911.02909 (2019)

Hot spot: non-equilibrium phonons



- Efficient Auger recombination
- Large energy release
- Excitation of non-equilibrium phonons

Phonons propagate out of the hot spot and drag excitons $\Rightarrow$ halo-like pattern is formed

Drift-diffusion model

$$\frac{\partial n}{\partial t} + \boldsymbol{\nabla} \cdot \boldsymbol{j} + \frac{n}{\tau} + R_A n^2 = 0,$$

$$\boldsymbol{j} = -D\boldsymbol{\nabla} n + \frac{\tau_p}{m}\boldsymbol{F}(\boldsymbol{\rho})n$$

$$\boldsymbol{F} = \boldsymbol{F}_{\text{phonons}} + \boldsymbol{F}_{\text{Seebeck}}$$

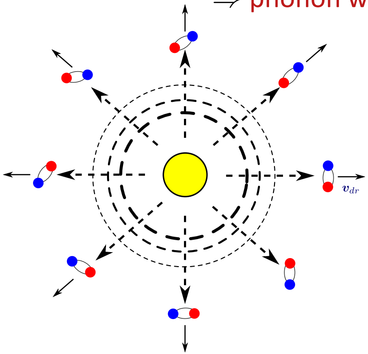
Phys. Rev. B **100**, 045426 (2019)

Phonon wind effect

Low temperatures, ballistic phonons

- Pump pulse creates hot spot
- ballistic phonons
- momentum flux

$\Rightarrow$ phonon wind



Phonons drag excitons away

Keldysh (1976); Zinov'ev, Ivanov, Kozub, Yaroshetskii (1983)
Bulatov, Tikhodeev (1992)

Force field produced by phonons (2D)
can be found from the kinetic equation

Exciton distribution function f_k:

$$\frac{\partial f_k}{\partial t} + v_k \frac{\partial f_k}{\partial r} + \frac{f_k - \bar{f}_k}{\tau_p} = -\frac{f_k}{\tau_d} + g_k$$

$$+Q_{\text{exc-ph}}\{f_k\}$$

At $f_k \ll 1$, and high phonon occupancies $N_q \gg 1$

$$Q_{\text{exc-ph}}\{f_k\} = \frac{2\pi}{\hbar}\sum_q |M_q|^2 (f_{k+q} - f_k) \times$$

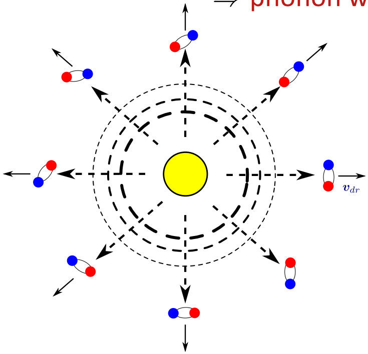
$$[N_q \delta(E_{k+q} - E_k - \hbar\Omega_q) + N_{-q}\delta(E_{k+q} - E_k + \hbar\Omega_q)]$$

Phonon wind effect

Low temperatures, ballistic phonons

- Pump pulse creates hot spot
- ballistic phonons
- momentum flux

$\Rightarrow$ phonon wind



Phonons drag excitons away

Keldysh (1976); Zinov'ev, Ivanov, Kozub, Yaroshetskii (1983)
Bulatov, Tikhodeev (1992)

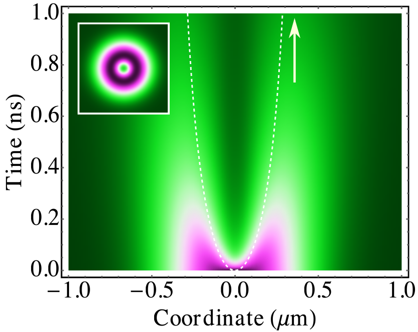
Force field produced by phonons (2D)

$$\boldsymbol{F}_{\rm wind}(\boldsymbol{\rho}) = \frac{U}{\rho}\,\frac{\boldsymbol{\rho}}{\rho}$$

Hot spot acts as a repulsive center

effective "Coulomb" repulsion

Cloud expansion $\rho(t) \approx \sqrt{Ut}$



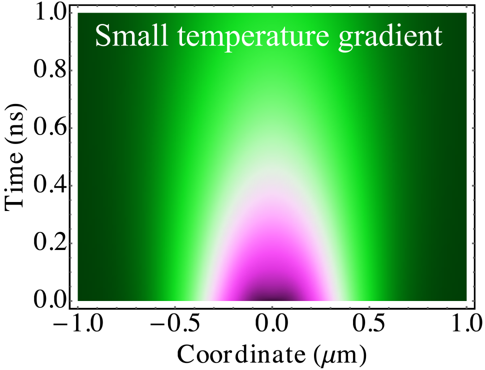
Phonon drag and Seebeck effects

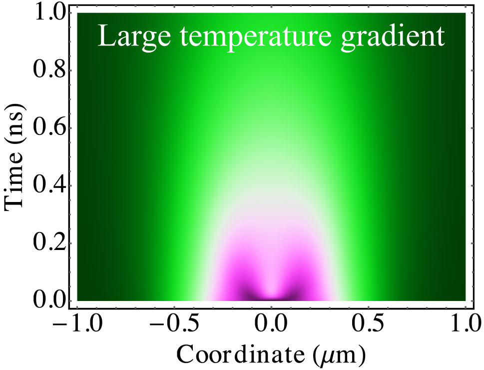
High temperatures, diffusive phonons

Temperature gradients of lattice and of excitons are formed

$$\boldsymbol{F}_{\text{drag}} = -\frac{\tau_p}{\tau_x} k_B \boldsymbol{\nabla} T_{\text{latt}}, \quad \boldsymbol{F}_{\text{Seebeck}} = -k_B \boldsymbol{\nabla} T_{\text{exc}}$$

Phonon drag scenario:





Phys. Rev. B **100**, 045426 (2019); **for Seebeck effect:** Perea-Causin et al. Nano Lett. **19**, 7317 (2019)

Phonon wind vs. phonon drag

Phonon wind

Phonons propagate ballistically

$$\boldsymbol{F}_{\text{wind}}(\boldsymbol{\rho}) = \frac{U}{\rho}\frac{\boldsymbol{\rho}}{\rho}$$

These bluestripe snapper are schooling. They are all swimming in the same direction in a coordinated way.

Phonon drag

Phonons propagate diffusively

$$\boldsymbol{F}_{\text{drag}}(\boldsymbol{\rho}) = \boldsymbol{\nabla}_{\rho}\frac{\Theta}{4\pi\varkappa t}\exp\left(-\frac{\rho^2}{4\varkappa t}\right)$$

These surgeonfish are shoaling. They are swimming somewhat independently, but in such a way that they stay connected.

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- Какие механизмы определяют коэффициент диффузии экситонов?
- Всегда ли применимо квазиклассическое описание?

Applicability of the quasi-classical description of exciton transport

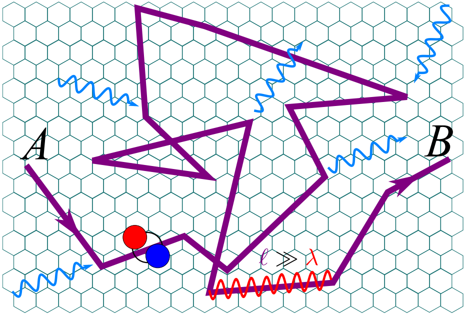
Non-degenerate excitons

$$\ell \gg \lambda \quad \Rightarrow \quad \frac{k_B T \tau}{\hbar} \gg 1$$

Diffusion coefficient

$$D = \left\langle \frac{v^2 \tau}{2} \right\rangle = \frac{k_B T \tau}{M} \gg \frac{\hbar}{M} \sim 1\ \mathrm{cm}^2/s,$$

otherwise quantum effects play role



LA-phonon scattering in MX_2 MLs

$$\tau = \frac{Ms^2}{k_B T}\tau_0, \quad \tau_0^{-1} = \frac{M^2(\Xi_c - \Xi_v)^2}{\rho \hbar^3} \quad \Rightarrow \quad D = s^2 \tau_0 \sim 1\ \mathrm{cm}^2/\mathrm{s}$$

In this mechanism D is temperature independent

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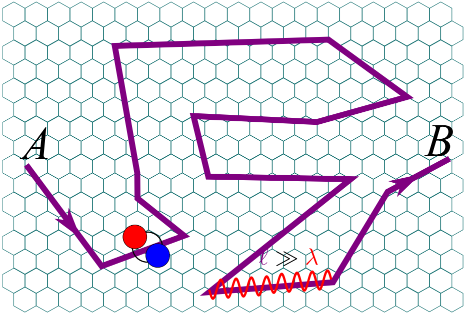

Our goal

is to study the lowest order corrections in the parameter $\hbar/(k_B T \tau) \ll 1$.

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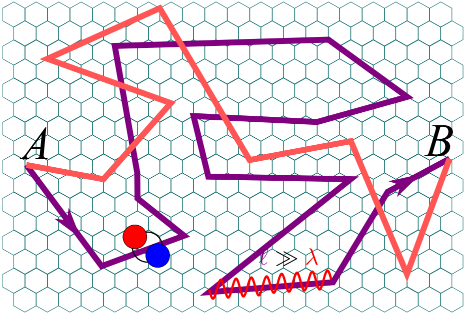

$$P(A \to B) = \left| \sum_i \mathcal{A}_i \right|^2, \quad \mathcal{A}_i = |\mathcal{A}_i| \exp\left(\mathrm{i}\phi_i\right)$$

$$\phi_i = \int_i \boldsymbol{k} \cdot d\boldsymbol{l}, \quad |\phi_i - \phi_j| \gtrsim k\ell \sim \frac{\ell}{\lambda} \gg 1 \quad \Rightarrow \quad P(A \to B) = \sum_i P_i \ (?)$$

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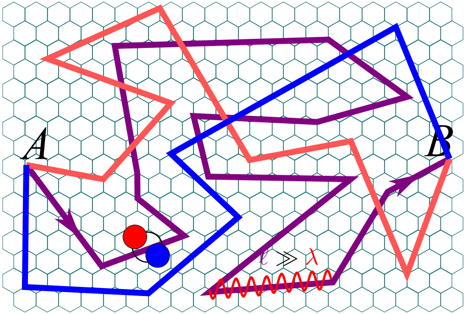

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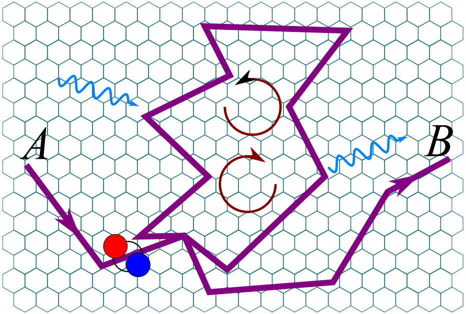

Diffusion coefficient

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otherwise quantum effects play role

Self-intersecting trajectory:

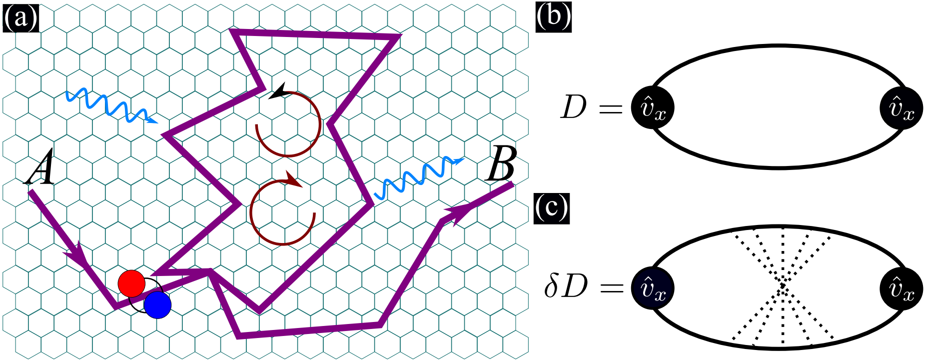
$$\phi_{\circlearrowright} = \phi_{\circlearrowleft}$$

Constructive interference $\Rightarrow$ localization

Exciton weak localization

arXiv:1911.10528 (2019)

$k_B T \gg Ms^2 \sim 1$ K   Quasi-elastic LA-phonon scattering



$$\frac{\delta D}{D} \sim -\int_{\tau}^{\tau_\phi} \frac{v_T \lambda dt}{Dt} \sim -\frac{\hbar}{k_B T \tau} \ln\left(\frac{\tau_\phi}{\tau}\right)$$

Exciton weak localization: Ivchenko, Pikus, Razbirin, Starukhin (1977), Arseev, Dzyubenko (1998)

Quasielasticity:

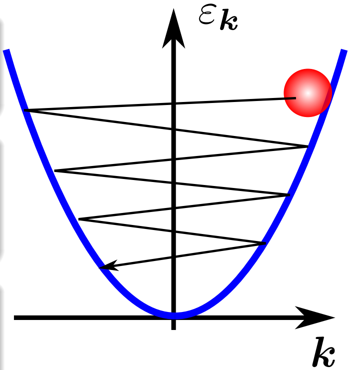
$$\Delta\epsilon \sim \sqrt{k_B T M s^2} \ll k_B T \quad \Rightarrow \quad \delta\epsilon^2(t) \sim (\Delta\epsilon)^2 \frac{t}{\tau}$$

Momentum relaxation time

$$\tau = \frac{Ms^2}{k_B T}\tau_0, \quad \tau_0^{-1} = \frac{M^2(\Xi_c - \Xi_v)^2}{\rho\hbar^3}$$

Energy relaxation time

$$\delta\epsilon(\tau_\epsilon) \sim k_B T \quad \Rightarrow \quad \tau_\epsilon = \frac{\tau_0}{2} \gg \tau$$



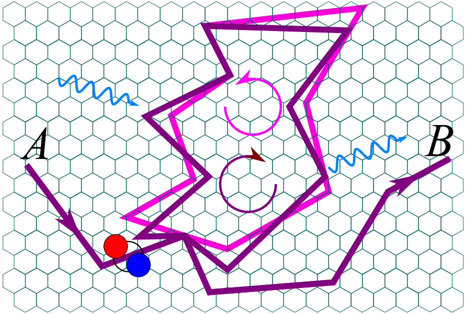
Phase relaxation time

$$\delta\epsilon(\tau_\phi) \sim \frac{\hbar}{\tau_\phi} \quad \Rightarrow \quad \tau_\phi \sim \left[\frac{\hbar^2\tau_0}{(k_B T)^2}\right]^{1/3} \quad \Rightarrow \quad \frac{\tau_\phi}{\tau} \propto T^{1/3}$$

Quantum effects become more pronounced with the temperature increase

Golubentsev (1984); Afonin, Galperin, Gurevich (1985); Adams, Paalanen (1987); Stephen (1987); Dyakonov, Kopelevich (1988)

Dephasing due to phonon propagation



$$C_\phi = \prod_i \exp\left\{ i \frac{\Delta \boldsymbol{p}_i}{\hbar} [\boldsymbol{r}(t_i) - \boldsymbol{r}(-t_i)] \right\}$$

$$\sim \exp\left\{ -\frac{M\epsilon^2}{\hbar^2 \tau} \int_{-t}^{t} [\boldsymbol{r}(t') - \boldsymbol{r}(-t')]^2 dt' \right\}$$

$$\sim \exp\left\{ -\frac{Ms^2\epsilon^2}{\hbar^2 \tau} \int_{-t}^{t} (t')^2 dt' \right\} \sim \exp\left[-\frac{t^3}{\tau_\phi^3(\varepsilon)} \right]$$

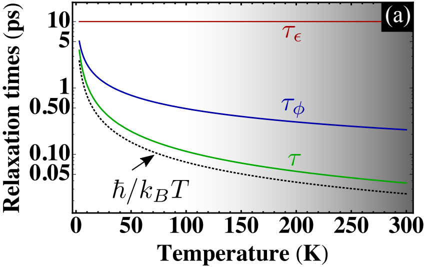
ballistic phonons: $r(t) \sim st$

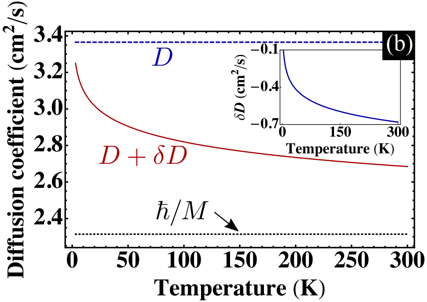
Phonons propagate with a constant velocity (s is the speed of sound), while the velocity of excitons increases with increasing the temperature.

Thus, dephasing is less efficient at elevated temperatures.

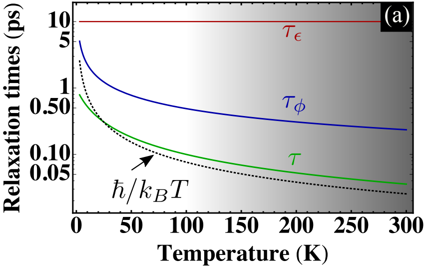
Weak localization: results

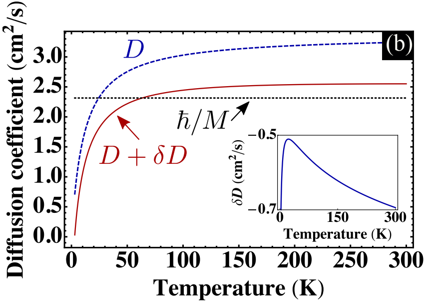
Phonons only





Phonons and impurities





arXiv:1911.10528 (2019)

Take home message

Excitons in 2D crystals are ideal objects to study linear and nonlinear transport effects.

Thank you for attention!

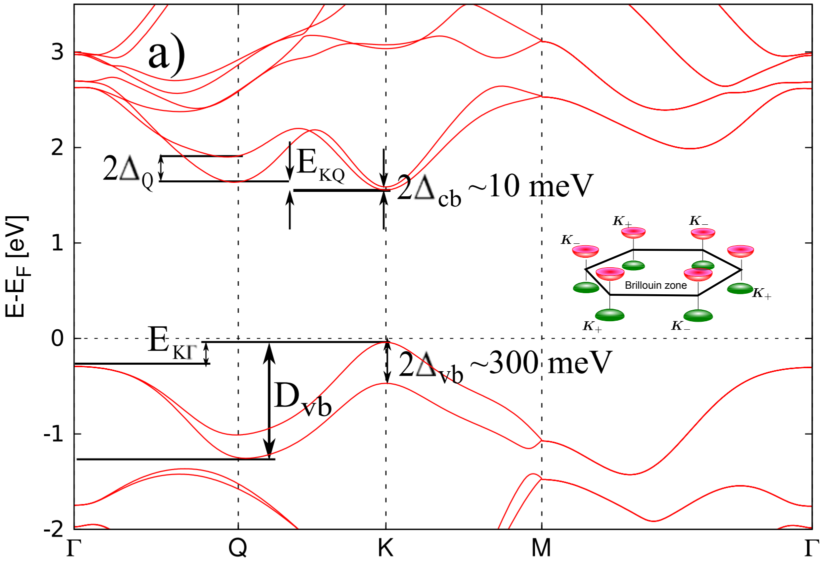
Kulig, ..., MMG, Chernikov, Exciton Diffusion and Halo Effects in Monolayer Semiconductors, Phys. Rev. Lett. **120**, 207401 (2018)

MMG, Phonon wind and drag of excitons in monolayer semiconductors, Phys. Rev. B **100**, 045426 (2019)

Zipfel, ..., MMG, Malic, Chernikov, Exciton diffusion in monolayer semiconductors with suppressed disorder, arXiv:1911.02909

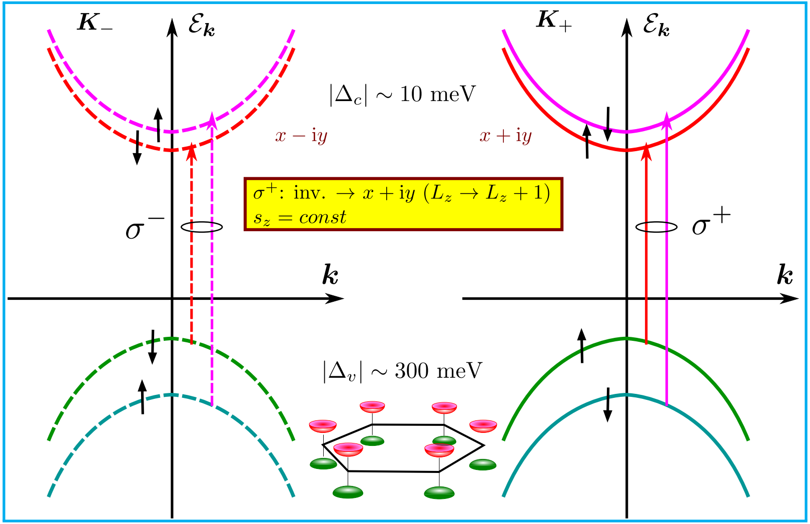
MMG, Quantum interference effect on exciton transport in monolayer semiconductors, arXiv:1911.10528

Electron spectrum



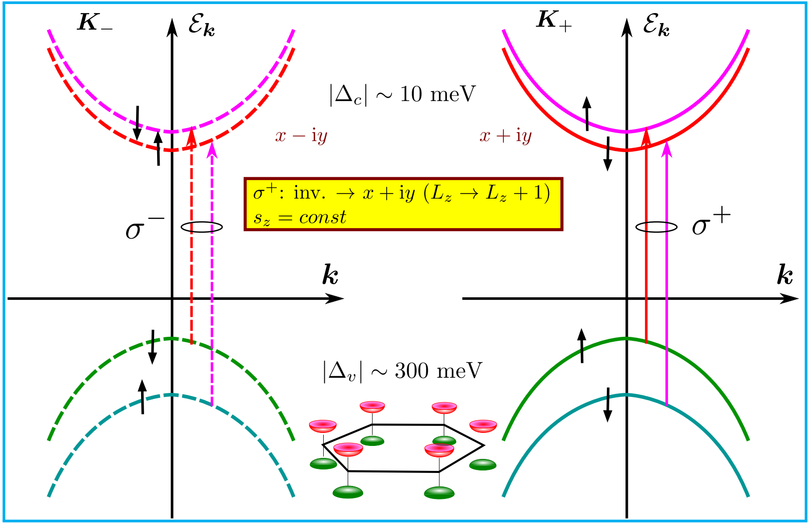
Kormanyos et al. (2015)

Mo-based



K_-
$\mathcal{E}_{\boldsymbol{k}}$
K_+
$\mathcal{E}_{\boldsymbol{k}}$
$|\Delta_c| \sim 10$ meV
$x - \mathrm{i}y$
$x + \mathrm{i}y$
σ^+: inv. $\rightarrow x + \mathrm{i}y$ $(L_z \rightarrow L_z + 1)$
$s_z = const$
σ^-
σ^+
$\boldsymbol{k}$
$\boldsymbol{k}$
$|\Delta_v| \sim 300$ meV

W-based



K_-
$\mathcal{E}_{\boldsymbol{k}}$
K_+
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$|\Delta_c| \sim 10$ meV
$x - \mathrm{i}y$
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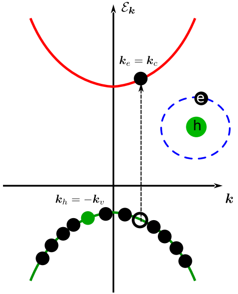
σ^+: inv. $\to x + \mathrm{i}y$ $(L_z \to L_z + 1)$
$s_z = const$
σ^-
σ^+
$\boldsymbol{k}$
$\boldsymbol{k}$
$|\Delta_v| \sim 300$ meV

Excitons in 2D semiconductors

Optically created electron-hole pairs form excitons

$$\Psi_{\mathrm{exc}} = \sum_{\boldsymbol{k}_e,\boldsymbol{k}_h} C_{\boldsymbol{k}_e,\boldsymbol{k}_h} |\boldsymbol{k}_e, s_e; \boldsymbol{k}_h, s_h\rangle = \sum_{\boldsymbol{k}_e,\boldsymbol{k}_h} C_{\boldsymbol{k}_e,\boldsymbol{k}_h} \mathcal{U}_\mu(\boldsymbol{k}_e, \boldsymbol{k}_h)$$

$|\boldsymbol{k}_e, s_e; \boldsymbol{k}_h, s_h\rangle$ is the wavefunction of the state where the $|\boldsymbol{k}_e, s_e\rangle$ conduction band state is occupied and the $\mathcal{K}|\boldsymbol{k}_h, s_h\rangle$ valence band state is empty



Exciton wavefunction

$$\Psi_{\boldsymbol{K};\nu,\mu}(\boldsymbol{\rho}_e, \boldsymbol{\rho}_h) = \frac{\exp(\mathrm{i}\boldsymbol{K}\boldsymbol{R})}{\sqrt{S}} \Phi_\nu(\boldsymbol{\rho}) \mathcal{U}_\mu(\boldsymbol{\rho}_e, \boldsymbol{\rho}_h)$$

Envelope function $(1s, 2p, \ldots)$

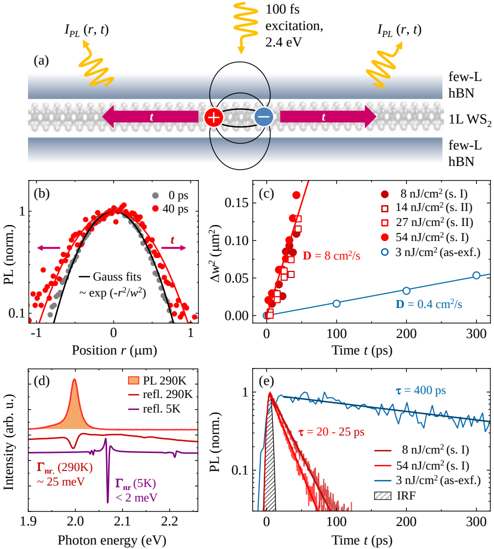
$$-\frac{\hbar^2}{2\mu}\Delta_\rho \Phi(\boldsymbol{\rho}) + V(\rho)\Phi(\boldsymbol{\rho}) = E\Phi(\boldsymbol{\rho})$$

non-parabolicity & SO-coupling: Trushin, Goerbig, Belzig (2018)

Encapsulated vs. non-encapsulated structures

hBN encapsulation suppresses the dielectric disorder

see, e.g., Archana Raja, …Alexey Chernikov, Dielectric disorder in two-dimensional materials, Nat. Nano. (2019)



arXiv:1911.02909 (2019)

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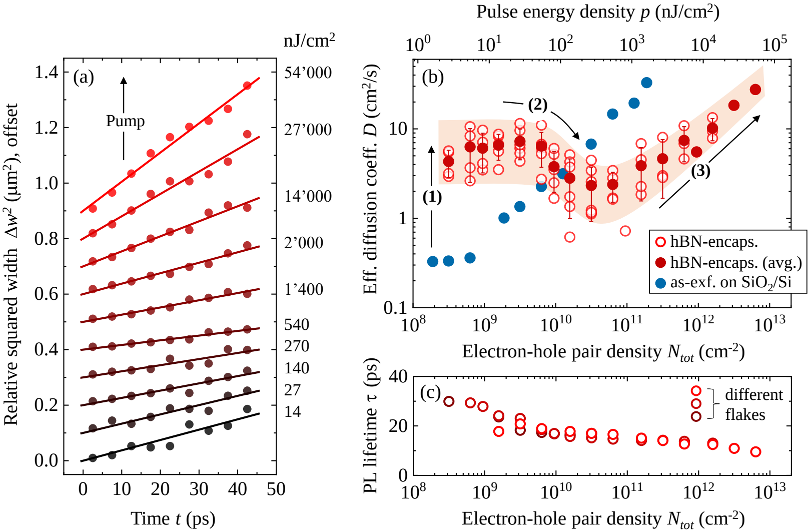
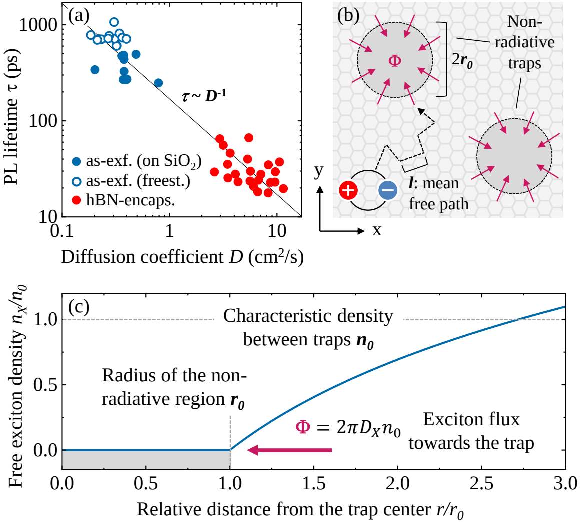


arXiv:1911.02909 (2019)

Diffusion limits the exciton lifetime

Correlation of the diffusion coefficient and the lifetime



Exciton density near traps

$$D\Delta n_X = 0$$

Flux to the trap

$$|\Phi| = 2\pi r_0 D_X \left.\frac{dn}{dr}\right|_{r=r_0}$$

Lifetime

$$\frac{1}{\tau_r} \equiv \frac{N_{tr}\Phi}{n_0} = 2\pi D_X N_{tr}$$

arXiv:1911.02909 (2019)

Electron-hole plasma vs. excitons

Binding energy

$$E_b \approx 200\ \text{meV}$$

Ionization equilibrium

between excitons and plasma

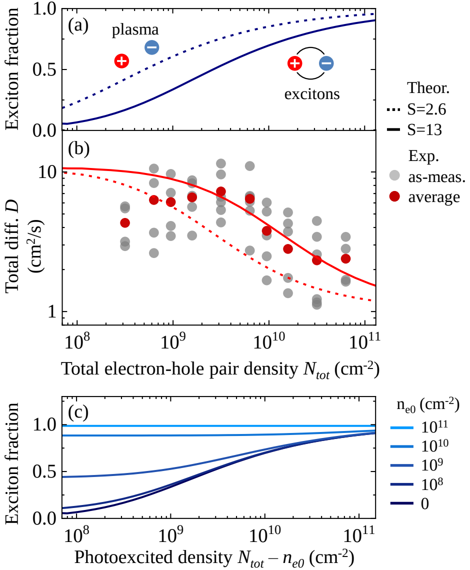
$$\frac{\bar{n}_X}{N_{tot}} = 1 + \frac{S}{2N_{tot}} - \sqrt{\left(\frac{S}{2N_{tot}}\right)^2 + \frac{S}{N_{tot}}}$$

Diffusion coefficients

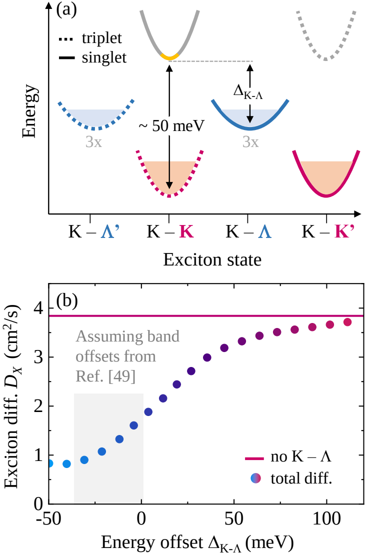
microscopic model (E. Malic et al.)

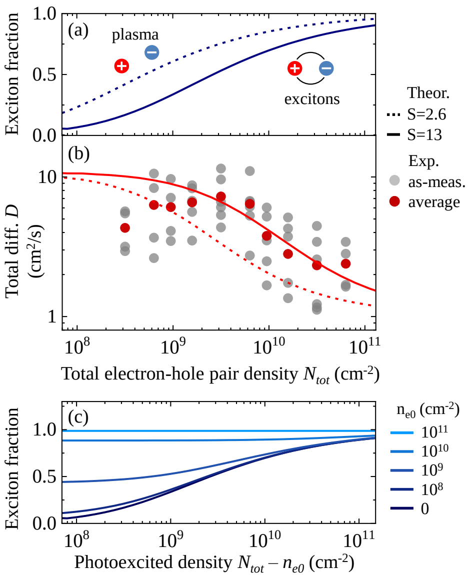
$$D_{eh} \approx 10\ \text{cm}^2/s;\ \ D_X \approx 1\ \text{cm}^2/s$$

arXiv:1911.02909 (2019)



Electron-hole plasma vs. excitons



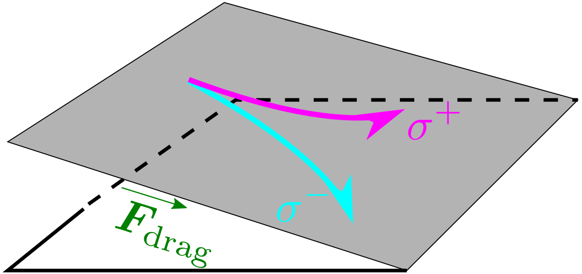


arXiv:1911.02909 (2019)

Valley Hall effect: separation of valleys

Valley Hall effect

Generation of the valley current transversal to the exciton flux $\quad \boldsymbol{i}_\sigma \propto [\boldsymbol{\sigma} \times \boldsymbol{F}_{\text{drag}}]$



Drag force $\boldsymbol{F}_{\text{drag}}$ can be caused by

- Inhomogeneous deformation
- Exciton temperature gradient (Seebeck effect)
- Phonon wind/drag

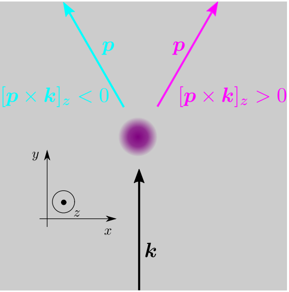
MMG (2019); Raül Perea-Causín et al. (2019)

In conventional systems

E. Hall (1881): AHE in ferromagnets
Karplus, Luttinger (1954); Smith (1955): theory
M.I. Dyakonov, V.I. Perel (1971): SHE prediction
Y. Kato (2004); J. Wunderlich (2005): observation

Microscopic theory: Three sources of the effect

Asymmetric scattering
(Mott effect, skew)

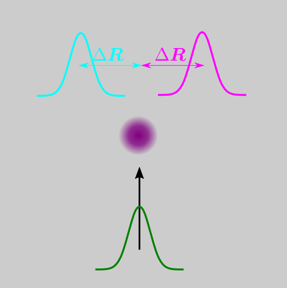


$$\boldsymbol{M}_{\boldsymbol{pk}} = M_q\left(1 + \mathrm{i}\xi[\boldsymbol{p}\times\boldsymbol{k}]_z\sigma_z\right)$$

$$\boldsymbol{i}_\sigma^{skew} = \frac{\xi}{\hbar}\frac{k_B T\tau}{\hbar}C_{as}[\boldsymbol{F}_{\mathrm{drag}}\times\hat{\boldsymbol{z}}]N$$

$$|C_{as}| \ll 1$$

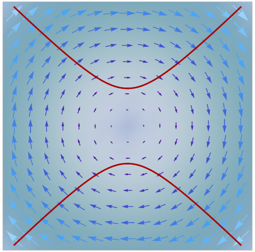
Shift of the wavepacket
(side-jump, shift)



$$\Delta\boldsymbol{R}_{\boldsymbol{pk}} \propto \xi[(\boldsymbol{p}-\boldsymbol{k})\times\hat{\boldsymbol{z}}]$$

$$\boldsymbol{i}_\sigma^{sj} \sim \frac{\xi}{\hbar}[\boldsymbol{F}\times\hat{\boldsymbol{z}}]N$$

Anomalous velocity
(intrinsic, Berry curvature)



$$\boldsymbol{v}_a \propto \xi[\boldsymbol{F}\times\hat{\boldsymbol{z}}]$$

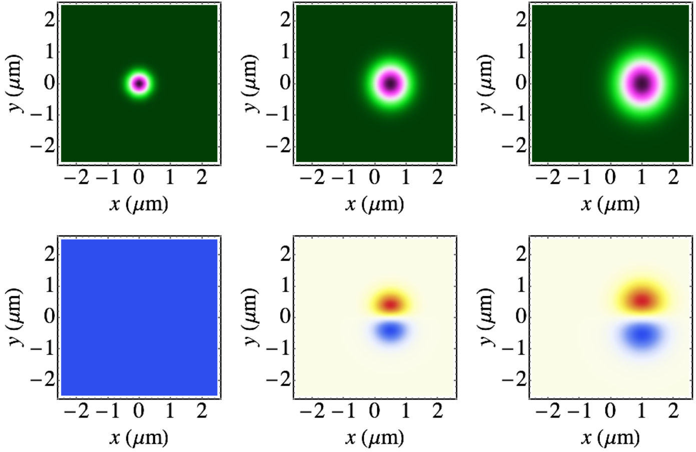
$$\boldsymbol{i}_\sigma^{a} \sim \frac{\xi}{\hbar}[\boldsymbol{F}\times\hat{\boldsymbol{z}}]N$$

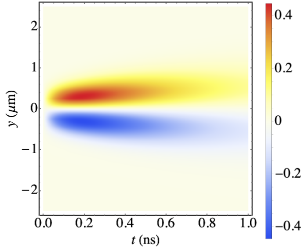
These contributions partially compensate each other

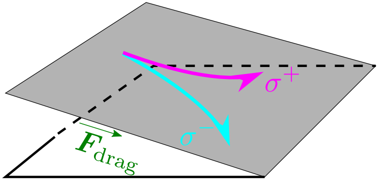
Exciton valley Hall effect: theoretical expectations

$$\frac{\partial n}{\partial t} = D\Delta n - \boldsymbol{f}_{\rm drag}\cdot\boldsymbol{\nabla} n - \zeta \boldsymbol{f}_{\rm drag}\cdot\boldsymbol{\nabla}\times\boldsymbol{S} - \frac{n}{\tau}, \quad \frac{\partial \boldsymbol{S}}{\partial t} = D\Delta\boldsymbol{S} - (\boldsymbol{f}_{\rm drag}\cdot\boldsymbol{\nabla})\boldsymbol{S} - \zeta\boldsymbol{f}_{\rm drag}\times\boldsymbol{\nabla} n - \frac{\boldsymbol{S}}{T_s}$$

$$\boldsymbol{f}_{\rm drag} = \frac{\tau}{M}\boldsymbol{F}_{\rm drag}, \quad \boldsymbol{S} = \sigma_z\hat{\boldsymbol{z}}$$







MMG, L.E. Golub, in preparation