

Экситоны в двумерных материалах

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ФТИ им. А.Ф. Иоффе, Санкт-Петербург

Лекция 2. Экситонный транспорт: классические и квантовые эффекты

- 1 Режимы экситонного транспорта
 - Прыжковый транспорт
 - Полуклассическое распространение
 - Слабая локализация экситонов
- 2 Нелинейный транспорт экситонов
 - Экситон-экситонные столкновения
 - Экситонные жидкости
- 3 Диффузия экситонов в море Ферми электронов

**ЛЕТНЯЯ ШКОЛА
ФОНДА «БАЗИС»**

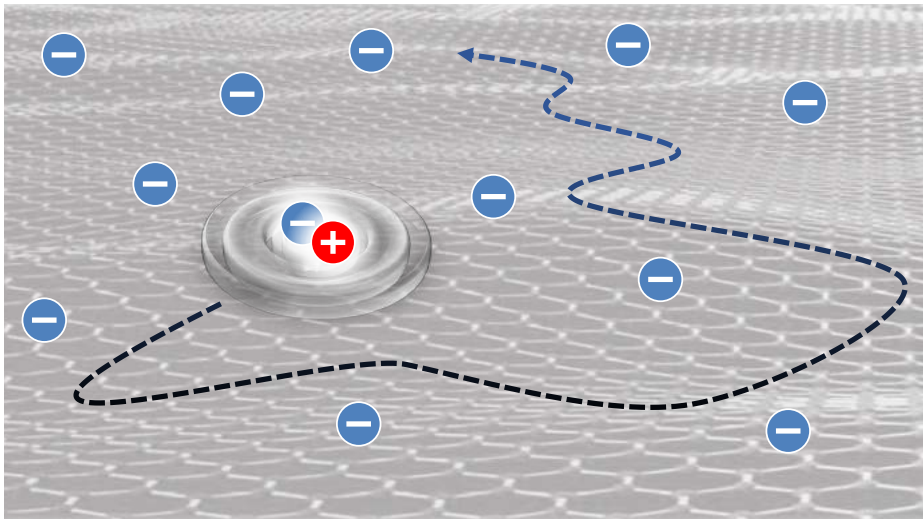
БАЗИС



Фонд «БАЗИС»
Санкт-Петербург, Россия

Exciton transport: what are we looking for?

Periodic crystal \Rightarrow Bloch theorem $\psi_{nk} = e^{ikr} u_{nk}(\mathbf{r}) \Rightarrow$ free propagation



static disorder (defects) + phonons + charge carriers + other excitons

How can we solve this very complex problem?

Phenomenological approach: macroscopic (coarse grain) description

R. Brown (1827); A. Einstein (1905); M. Smoluchowski (1906); N. Wiener (1918)

- Continuity equation for exciton density $n(\mathbf{r}, t)$

$$\frac{\partial n(\mathbf{r}, t)}{\partial t} + \text{div } \mathbf{i}(\mathbf{r}, t) = 0$$

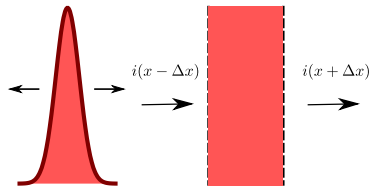
- Ficks law for exciton flux $\mathbf{i}(\mathbf{r}, t)$

$$\mathbf{i}(\mathbf{r}, t) = -D \nabla n(\mathbf{r}, t)$$

$D \geq 0$ is the diffusion coefficient

~ expansion in small gradient of density

Density should be smooth on the microscopic length and timescales



Diffusion equation allowing for the generation, G , and recombination, R

$$\frac{\partial n}{\partial t} - D \Delta n + R\{n\} = G$$

Linear diffusion equation

$$\frac{\partial n}{\partial t} - D\Delta n + \frac{n}{\tau} = G(\mathbf{r}, t)$$

General solution in 2D:

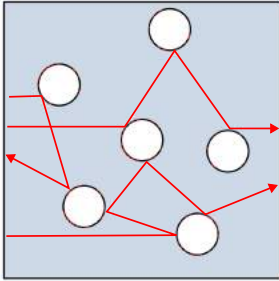
$$n(\mathbf{r}, t) = \int_{-\infty}^t dt' \int d\mathbf{r}' g(\mathbf{r} - \mathbf{r}', t - t') e^{-\frac{t-t'}{\tau}} G(\mathbf{r}', t'), \quad g(\mathbf{r}, t) = \frac{1}{4\pi Dt} \exp\left(-\frac{r^2}{4Dt}\right)$$

The mean squared displacement along x or y

$$\sigma^2(t) = \int dx x^2 g(x, t) = \int dy y^2 g(y, t) = 2Dt$$

$$\text{displacement } \sigma \propto \sqrt{t}$$

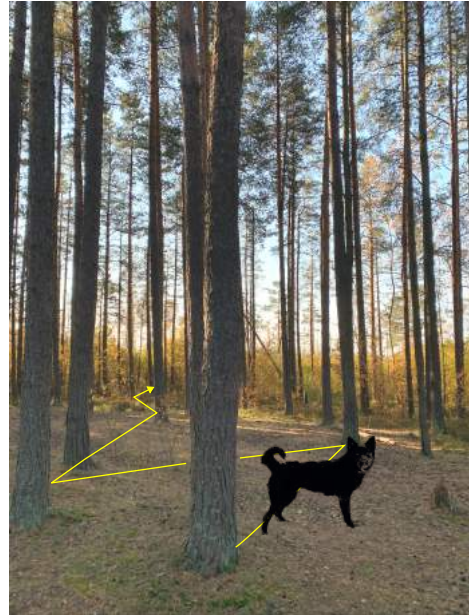
Diffusion and random walk: Free propagation interrupted by scattering



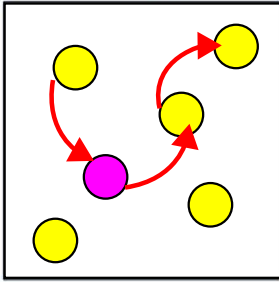
$$x^2(t) = \left(\sum_i \Delta x_i \right)^2 = \underbrace{\sum_i (\Delta x_i)^2}_{\sim \ell^2 n_{\text{steps}}} + \underbrace{\sum_{i \neq j} \Delta x_i \Delta x_j}_{=0} = \ell^2 \frac{t}{\tau_p}$$

mean free path $\ell = v\tau_p$, scattering time τ_p , $n_{\text{steps}} \approx t/\tau_p$

diffusion coefficient $D = \frac{v^2 \tau_p}{2}$



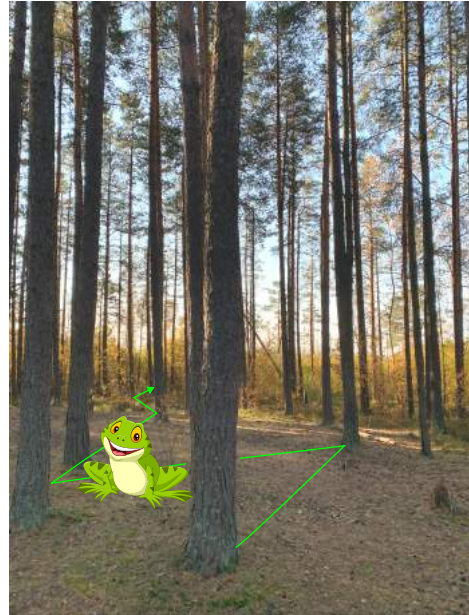
Diffusion and random walk: Hopping between localization sites



$$x^2(t) = \left(\sum_i \Delta x_i \right)^2 = \underbrace{\sum_i (\Delta x_i)^2}_{\sim d^2 n_{\text{steps}}} + \underbrace{\sum_{i \neq j} \Delta x_i \Delta x_j}_{=0} = d^2 \frac{t}{\tau_h}$$

hopping distance d , hopping time τ_h , $n_{\text{steps}} \approx t / \tau_h$

diffusion coefficient $D = \frac{d^2}{2\tau_h}$



Diffusion coefficient is determined by velocity correlations

General approach (valid in any transport regime):

$$x(t) = \int_0^t v_x(t_1) dt_1$$

We calculate the mean squared displacement

$$\langle x^2(t) \rangle = \left\langle \int_0^t v_x(t_1) dt_1 \int_0^t v_x(t_2) dt_2 \right\rangle = \int_0^t dt_1 \int_0^t dt_2 \langle v_x(t_1) v_x(t_2) \rangle$$

Velocity autocorrelation function

$$\langle v_x(t) v_x(t + \tau) \rangle = \lim_{T \rightarrow \infty} \frac{1}{T} \int_{-T/2}^{T/2} v_x(t) v_x(t + \tau)$$

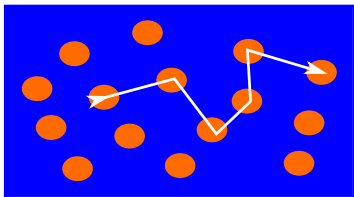
depends on the absolute difference of times $\tau = |t_2 - t_1|$

$$\langle x^2(t) \rangle = 2t \int_0^t \langle v_x(0) v_x(\tau) \rangle d\tau \xrightarrow[t \rightarrow \infty]{} 2Dt$$

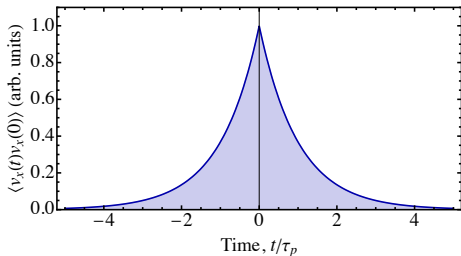
diffusion coefficient $D = \int_0^\infty \langle v_x(0) v_x(\tau) \rangle d\tau$

Velocity autocorrelation function: example

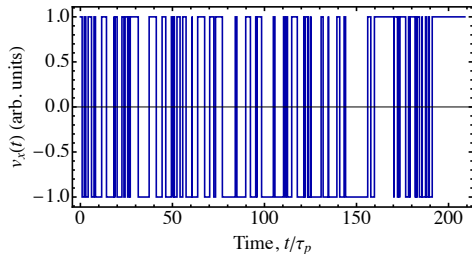
diffusion coefficient $D = \int_0^\infty \langle v_x(t)v_x(0) \rangle dt$



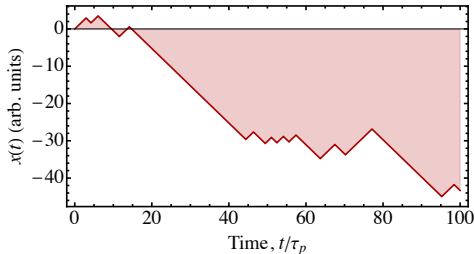
$$\langle v_x(t)v_x(0) \rangle \propto \exp(-|t|/\tau_p)$$



Velocity:



Coordinate:



Drift and diffusion: The Einstein relation

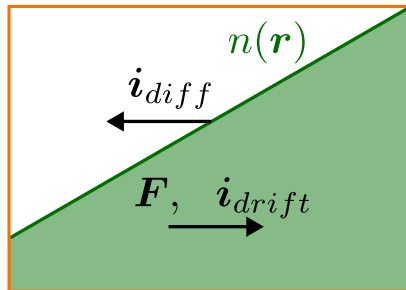
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- Ficks law for exciton flux $\mathbf{i}(\mathbf{r}, t)$

$$\mathbf{i}(\mathbf{r}, t) = -D \nabla n(\mathbf{r}, t)$$

$D \geq 0$ is the diffusion coefficient



Let an external force F create the **drift** exciton flux \mathbf{i} :

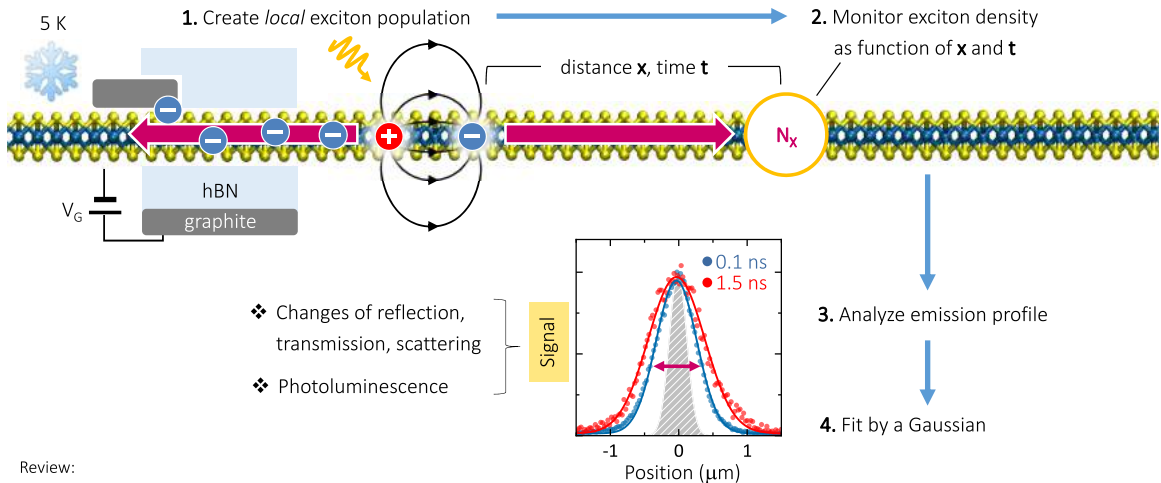
$$\mathbf{i}_{drift} = n \mathbf{v}_{dr} = n \mu \frac{\mathbf{F}}{|e|}, \quad \mu \text{ is the effective mobility}$$

In bounded system $\mathbf{i}_{tot} = \mathbf{i}_{drift} + \mathbf{i}_{diff} = 0$

the Einstein relation $\mu k_B T = |e| D$

It is a simplest example of the **fluctuation-dissipation theorem**

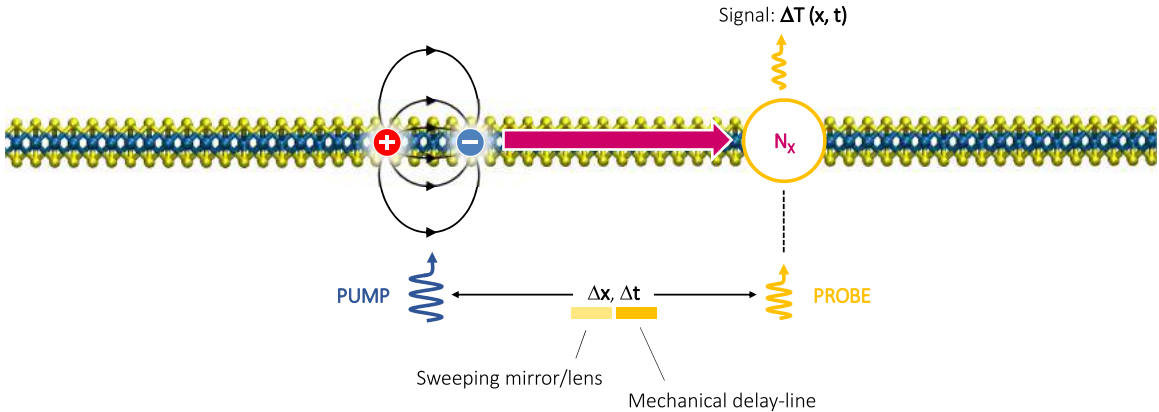
How to measure exciton propagation?



Review:

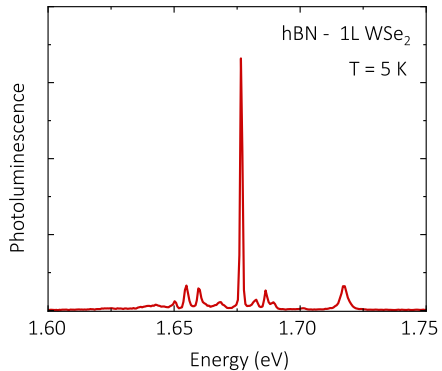
N. S. Ginsberg & W. A. Tisdale, *Annu. Rev. Phys. Chem.* 70 (2020)

(differential) absorption-based measurements

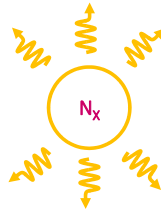


luminescence-based measurements

Detecting emitted light

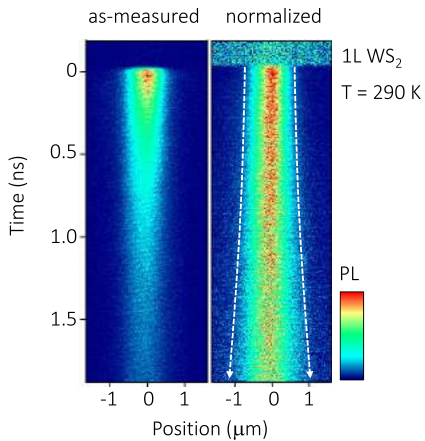


Signal: $PL(x, t)$

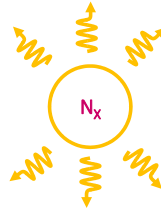


- ❖ Imaging of luminescence
- ❖ Time-resolved detection

luminescence-based measurements

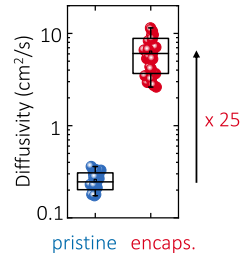
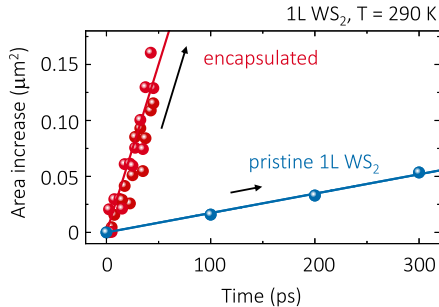
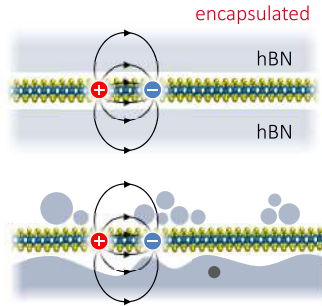


Signal: $\text{PL}(\mathbf{x}, t)$



- ❖ Imaging of luminescence
- ❖ Time-resolved detection

Influence of environment / disorder



Dielectric disorder

$$\epsilon(x) \neq \text{const.}$$

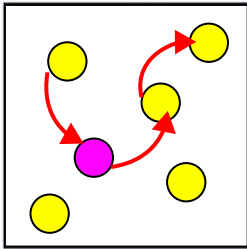
$$\mu_{\text{eff}} = De/k_B T$$

*assuming semiclassics $D = \frac{k_B T \tau_s}{M_X}$

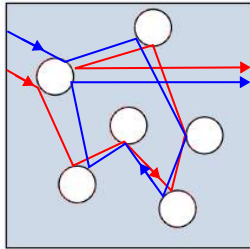
❖ Effective mobility up to 400 cm²/Vs - 20 nm mean free path*, 100's nm diffusion length

Regimes of exciton propagation

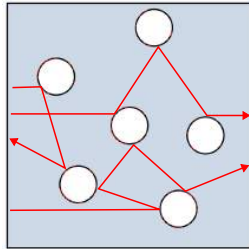
a hopping



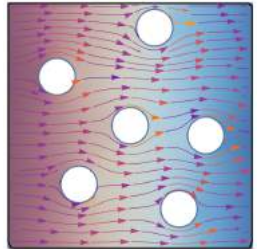
b weak localization



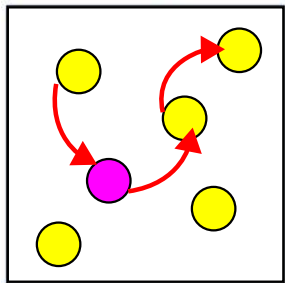
c semiclassical



d hydrodynamic



Exciton hopping between localization sites



hopping distance d , hopping time τ_h , $n_{\text{steps}} \approx t/\tau_h$:

diffusion coefficient $D = \frac{d^2}{2\tau_h} \propto \exp\left[-\left(\frac{T_0}{T}\right)^s\right]$

$s = 1/3$ for the variable range hopping in 2D

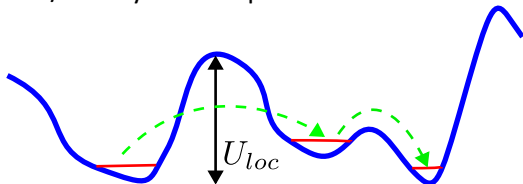
$$k_B T_0 \sim U_{loc}$$

Hopping transport can be very complicated and very slow

Hopping is realized if excitons are localized in disorder potential

$$U_{loc} \gtrsim k_B T,$$

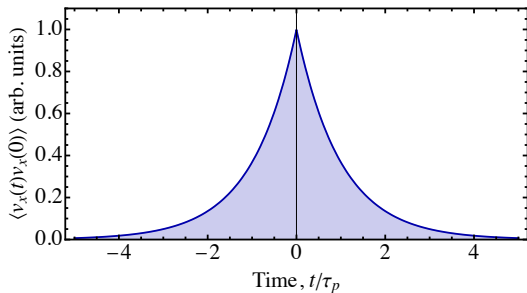
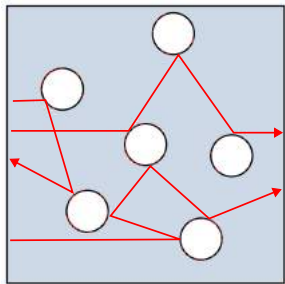
e.g., moire potentials in heterstructures and/or very low temperatures



- Exponential temperature dependence
 $T \uparrow \Rightarrow D \uparrow$
- $\sigma(t)$ can be non-diffusive due to spread in d and τ_h

Semiclassical propagation: basics

$$\text{applicability: } k_B T \gg U_{loc} \Leftrightarrow \frac{k_B T \tau_p}{\hbar} \gg 1 \Leftrightarrow \ell_{\text{mfp}} = v \tau_p \gg \lambda_{\text{de Broglie}} \sim \sqrt{\frac{\hbar^2}{M k_B T}}$$



$$D = \int_0^{\infty} \langle v_x(t)v_x(0) \rangle dt$$

$$\langle v_x(t)v_x(0) \rangle = \langle v_x^2(0) \rangle \exp(-|t|/\tau_p)$$

$$\text{non-degenerate excitons} \quad \left\langle \frac{Mv_x^2}{2} + \frac{Mv_y^2}{2} \right\rangle = k_B T \Rightarrow \langle v_x^2(0) \rangle = \frac{k_B T}{M}$$

$$\text{diffusion coefficient} \quad D = \frac{k_B T}{M} \tau_p$$

Kinetic equation: rigorous approach. Beyond coarse grain description

$$\text{applicability: } k_B T \gg U_{loc} \Leftrightarrow \frac{k_B T \tau_p}{\hbar} \gg 1 \Leftrightarrow \ell_{\text{mfp}} = v \tau_p \gg \lambda_{\text{de Broglie}} \sim \sqrt{\frac{\hbar^2}{M k_B T}}$$

Exciton distribution function $f_{m,k}(t)$ obeys kinetic equation

$$\frac{\partial f_{m,k}}{\partial t} + \underbrace{v_{m,k} \cdot \frac{\partial f_{m,k}}{\partial \mathbf{r}}}_{\text{propagation}} + \underbrace{\frac{\mathbf{F}}{\hbar} \cdot \frac{\partial f_{m,k}}{\partial \mathbf{k}}}_{\text{drift}} + \underbrace{\hat{Q}\{f_{m,k}\}}_{\text{scattering}} + \underbrace{\hat{R}\{f_{m,k}\}}_{\text{recombination}} = \underbrace{G_{m,k}(\mathbf{r}, t)}_{\text{generation}}$$

m enumerates bands, k is the wavevector.

$$\text{Collision integral } \hat{Q}\{f_{m,k}\} = \sum_{m',k'} W_{m,k;m',k'} f_{m',k'} - W_{m',k';m,k} f_{m,k}$$

Single band, effective mass model $E_k = \hbar^2 k^2 / 2M$, (quasi)elastic scattering:

$$\text{diffusion coefficient } D = \frac{k_B T}{M} \tau_p, \quad \frac{1}{\tau_p} = \sum_{k'} W_{k,k'} (1 - \cos \vartheta_{k,k'})$$

Semiclassical regime works where excitons are well-defined quasiparticles/wavepackets

Key scattering mechanisms

Single band, effective mass model $E_k = \hbar^2 k^2 / 2M$, (quasi)elastic scattering:

$$\text{diffusion coefficient } D = \frac{k_B T}{M} \tau_p, \quad \frac{1}{\tau_p} = \frac{2\pi}{\hbar} \sum_{k'} |M_{k'k}|^2 \delta(E_k - E_{k'}) (1 - \cos \vartheta_{k,k'})$$

Static defects

$$U(\mathbf{r}) = \sum_i V_0 \delta(\mathbf{r} - \mathbf{R}_i)$$

$$|M_{k'k}|^2 = V_0^2 n_d \Rightarrow \tau_p = \text{const}(T)$$

Diffusion coefficient increases with T :

$$D \propto T$$

Phonons (long wavelength acoustic)

$$U(\mathbf{r}, t) = \sum_q \sqrt{\frac{\hbar}{2\rho\Omega_q}} i q \Xi \hat{b}_q e^{i q \mathbf{r} - i \Omega_q t} + \text{h.c.}$$

$$\tau_p^{-1} \propto T$$

Diffusion coefficient is T -independent:

$$D = \text{const}(T)$$

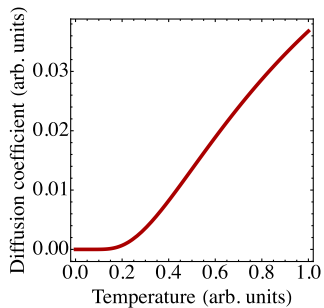
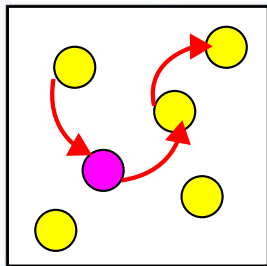
details

Selig et al., *Nat. Commun* **7**, 13279 (2016); S. Shree, et al., *PRB* **98**, 035302 (2018); MMG, *PRL* **124**, 166802 (2020); MMG et al., *APL* **121**, 192106 (2022)

These are simplest models, the reality is more complex

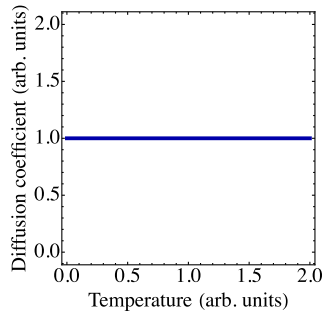
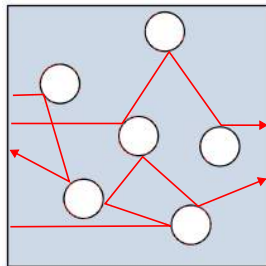
Summary of exciton propagation

hopping



D strongly increases with increase in T

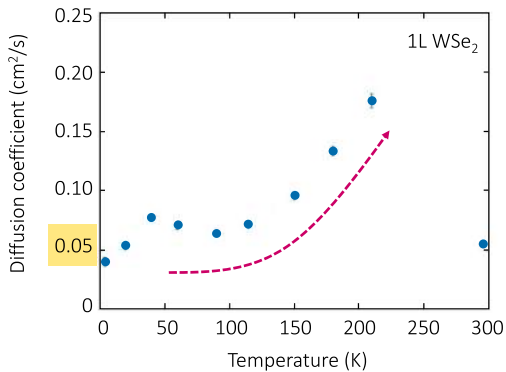
semiclassical



D weakly depends on T

Hopping

thermally activated diffusion

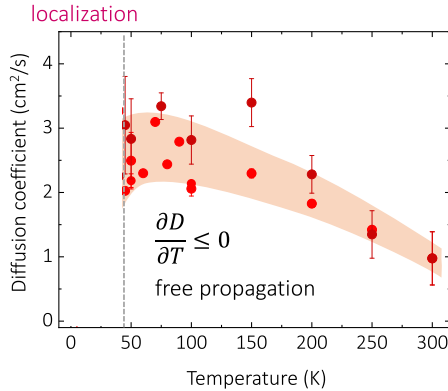
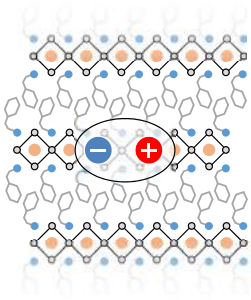


[P. Deotare lab]

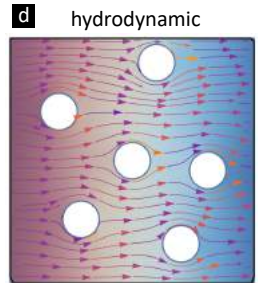
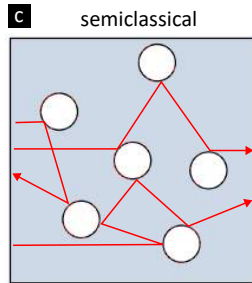
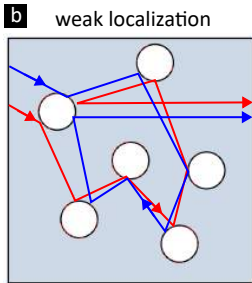
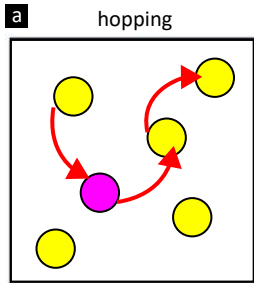
Z. Li et al, *ACS Nano* 15, 1539 (2021)

Free propagation

thermally “suppressed” diffusion

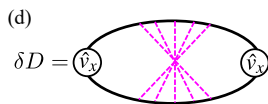
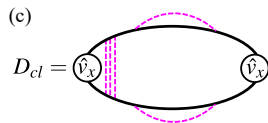
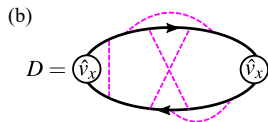
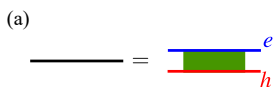


Regimes of exciton propagation



General approach is to determine D from exciton propagator $D = \int_0^\infty \langle\langle \hat{v}_x(t) \hat{v}_x(0) \rangle\rangle dt$

$$D = \frac{\hbar \sum_k \int_{-\infty}^{\infty} \frac{d\varepsilon}{2\pi} \text{Tr}\{\hat{n}_k \langle \hat{v}_x \hat{\mathcal{G}}_k^R(\varepsilon) \hat{v}_x \hat{\mathcal{G}}_k^A(\varepsilon) \rangle\}}{\sum_k \text{Tr}\{\hat{n}_k\}}$$



It is impossible to calculate and sum up all diagrams. In some cases, only specific ones contribute.

- Ladder (c) at the semiclassical regime

Do we need to go beyond semiclassics?

- Check diagram (d)!

Quantum interference of pathways

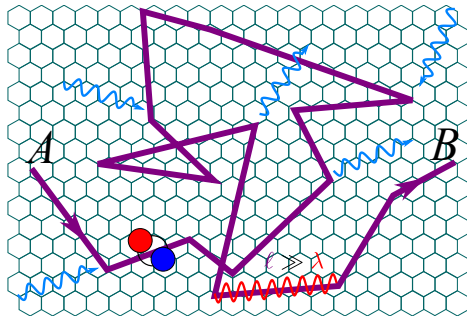
$$D = \int_0^\infty \langle\langle \hat{v}_x(t) \hat{v}_x(0) \rangle\rangle dt$$

Non-degenerate excitons: $l \gg \lambda \Rightarrow \frac{k_B T \tau}{\hbar} \gg 1$

semiclassical diffusion coefficient from

$$\langle v_x(t) v_x(0) \rangle = v_x^2(0) e^{-t/\tau}, \quad \frac{1}{\tau} = \sum_{k'} W_{kk'} (1 - \cos \vartheta)$$

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



LA-phonon scattering in MX₂ MLs

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

Here D is temperature independent; experiment at 4 K gives similar values ($\sim 2.5 \text{ cm}^2/\text{s}$).

Nonclassical effects should play a role

Quantum interference of pathways

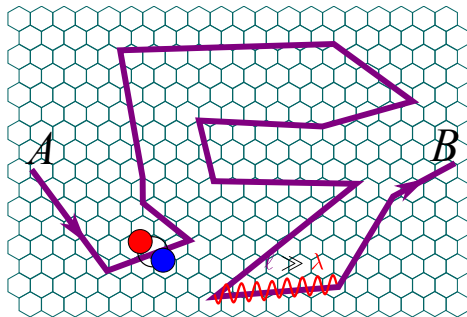
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$$D = \left\langle \frac{v^2 \tau}{2} \right\rangle = \frac{k_B T \tau}{M} \gg \frac{\hbar}{M} \sim 1 \text{ cm}^2/\text{s},$$



Quantum mechanics: different trajectories should interfere

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

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

Quantum interference of pathways

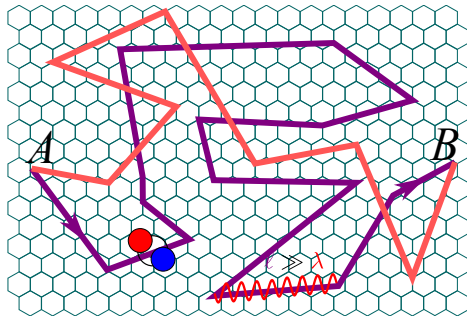
$$D = \int_0^\infty \langle\langle \hat{v}_x(t) \hat{v}_x(0) \rangle\rangle dt$$

Non-degenerate excitons: $\ell \gg \lambda \Rightarrow \frac{k_B T \tau}{\hbar} \gg 1$

semiclassical diffusion coefficient from

$$\langle v_x(t) v_x(0) \rangle = v_x^2(0) e^{-t/\tau}, \quad \frac{1}{\tau} = \sum_{k'} W_{kk'} (1 - \cos \vartheta)$$

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



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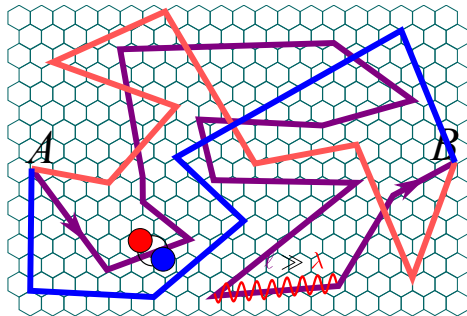
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Quantum interference of pathways

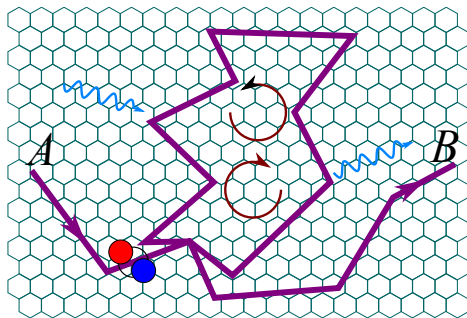
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semiclassical diffusion coefficient from

$$\langle v_x(t) v_x(0) \rangle = v_x^2(0) e^{-t/\tau}, \quad \frac{1}{\tau} = \sum_{k'} W_{kk'} (1 - \cos \vartheta)$$

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



Self-intersecting trajectory: $\oint k dl = \oint (-k) d(-l) = \oint k dl$

Constructive interference at perfectly elastic scattering \Rightarrow **localization**

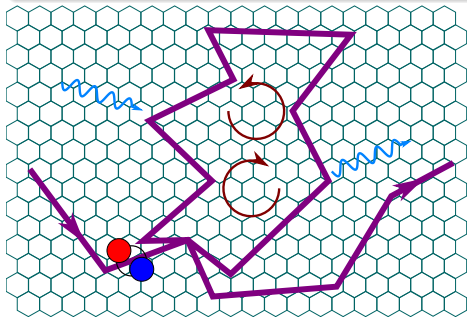
How well do the phases match in the case of the exciton-phonon scattering?

Non-trivial temperature dependence of the interference contribution

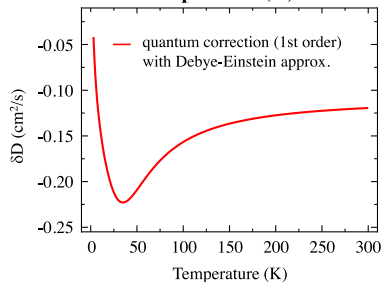
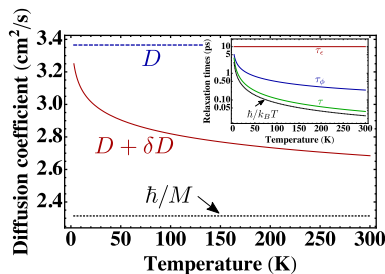
bulk crystals: Ivchenko, Pikus, Razbirin, Starukhin (1977); Golubentsev (1984); Afonin, Galperin, Gurevich (1985)

Acoustic phonon scattering is the main exciton scattering mechanism in MX_2 MLs at $1 \text{ K} \lesssim T \lesssim 50 \text{ K}$, $\tau \propto T^{-1}$:

$$D_{cl} = \frac{k_B T}{M} \tau = \text{const}(T)$$



$$\delta D = -\frac{\hbar}{2\pi M} \ln\left(\frac{\tau_\phi}{\tau}\right), \quad \frac{\hbar}{\tau_\phi} = \Delta\varepsilon(\tau_\phi), \quad \frac{\tau_\phi}{\tau} \propto T^{1/3}$$

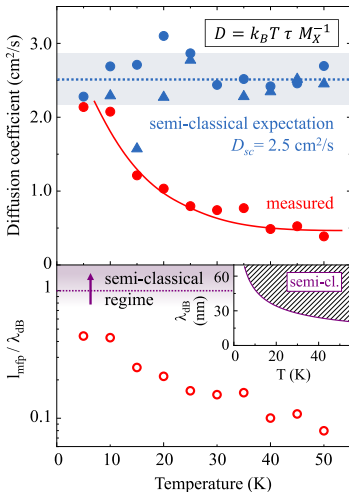
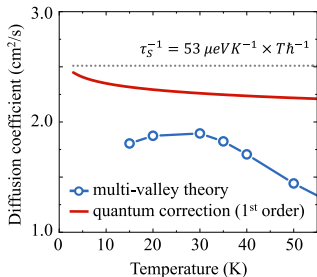


details

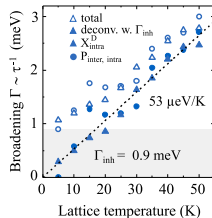
Semiclassical approach $D = \left\langle \frac{v^2 \tau}{2} \right\rangle, \quad \frac{1}{\tau} = \frac{2}{\hbar} \text{Im} \Sigma(E_k^x, \mathbf{k})$

Scattering contribution to the linewidth $\Gamma = \frac{2}{\hbar} \text{Im} \Sigma(0, 0) \left(= \frac{1}{\tau} \right)$

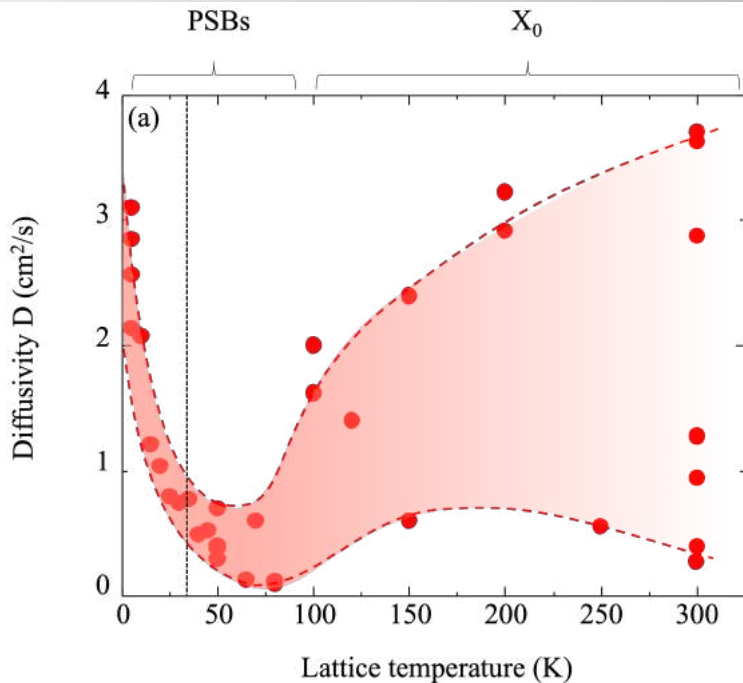
Theoretical expectations



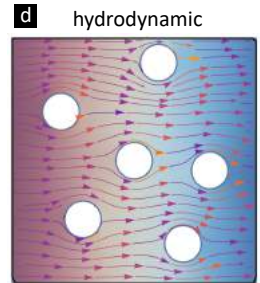
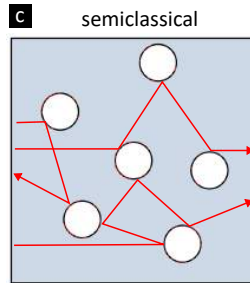
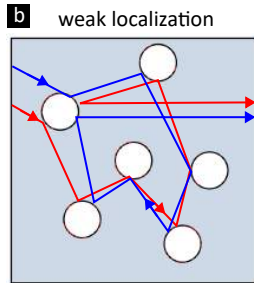
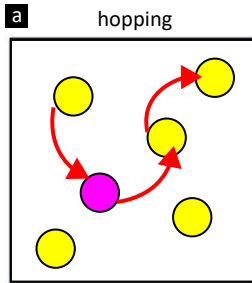
T-dependent linewidth



consistent with acoustic phonon scattering



Regimes of exciton propagation



What happens at elevated exciton densities and more extreme conditions?

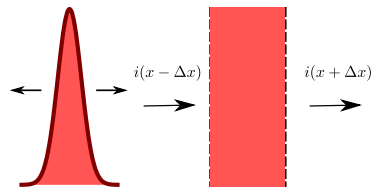
Diffusion in the presence of nonlinearities and external perturbations

- Continuity equation for exciton density $n(\mathbf{r}, t)$

$$\frac{\partial n(\mathbf{r}, t)}{\partial t} + \text{div } \mathbf{i}(\mathbf{r}, t) + R\{n\} = 0$$

- Ficks law for exciton flux $\mathbf{i}(\mathbf{r}, t)$

$$\mathbf{i}(\mathbf{r}, t) = -D\nabla n(\mathbf{r}, t) + \frac{D}{k_B T} n \mathbf{F}, \quad \mathbf{F} = -\nabla V$$



Nonlinearities

- Nonlinear recombination (Auger-like **exciton-exciton annihilation**): $R\{n\} = \frac{n}{\tau} + R_A n^2$
- **Exciton-exciton repulsion**: $\mathbf{F} = -U_0 \nabla n$
- **Temperature gradients** (feedback from EEA): $\mathbf{F} \propto -\nabla T$
- Screening, instabilities, etc., ... $D(n)$

Nonlinearities cause an increase of the **effective diffusion coefficient**

Nonlinear diffusion equation

$$\frac{\partial n}{\partial t} + \frac{n}{\tau} + R_A n^2 = D \Delta n + \frac{U_0 D}{k_B T} \nabla \cdot (n \nabla n) + \frac{D}{k_B T} \nabla \cdot (n \nabla V)$$

Gaussian initial condition

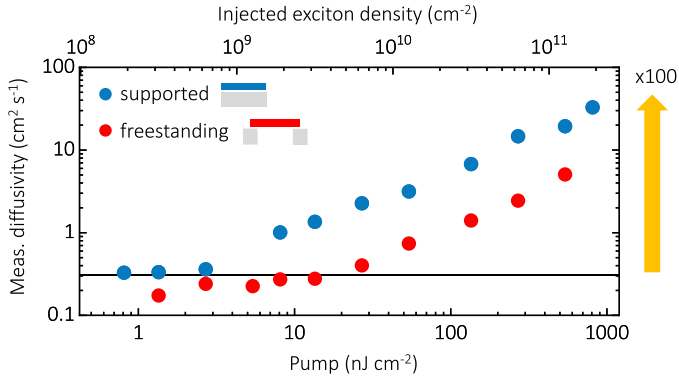
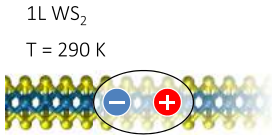
$$n(\mathbf{r}, 0) = \frac{N_0}{\pi r_0^2} e^{-r^2/r_0^2}$$

Effective diffusion coefficient

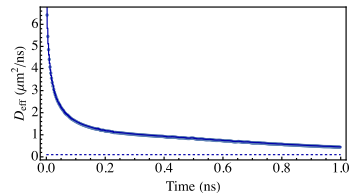
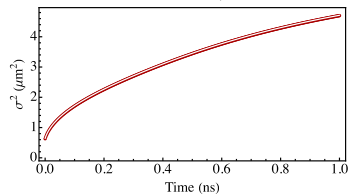
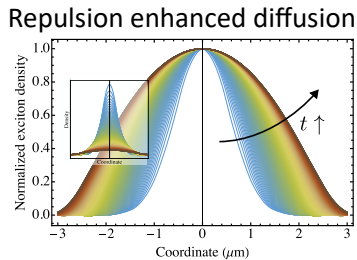
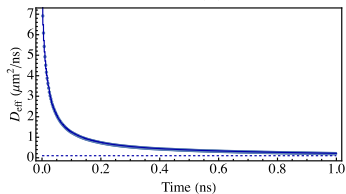
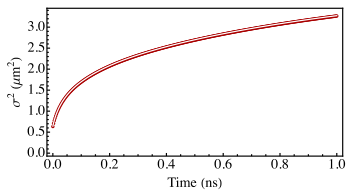
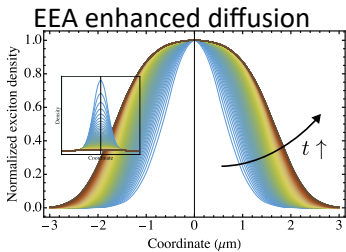
$$D_{\text{eff}} = -\frac{\pi r_0^4}{4N_0} \left[N_0 \frac{\partial}{\partial t} \frac{n}{N_0} \right]_{r=0, t=0} = D + R_A \frac{N_0}{8\pi} + \frac{U_0 D}{k_B T} \frac{N_0}{\pi r_0^2} + \frac{D}{k_B T} \frac{r_0^2}{4} \Delta V|_{r=0}$$

Note that D is only weakly renormalized by XX scattering due to momentum conservation

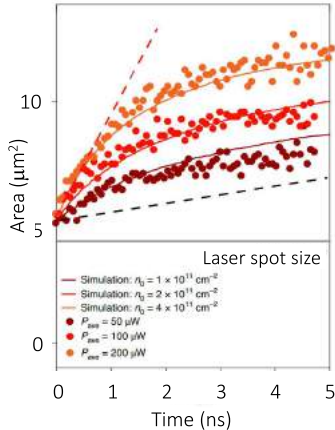
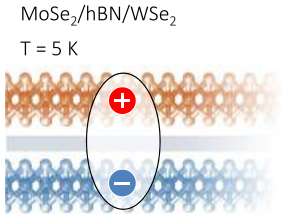
Non-linear diffusion



Nonlinearities modify exciton density profile yielding $D_{\text{eff}} \neq D$



Non-linear diffusion in TMDC heterobilayers



Density dependence

Subdiffusive behavior

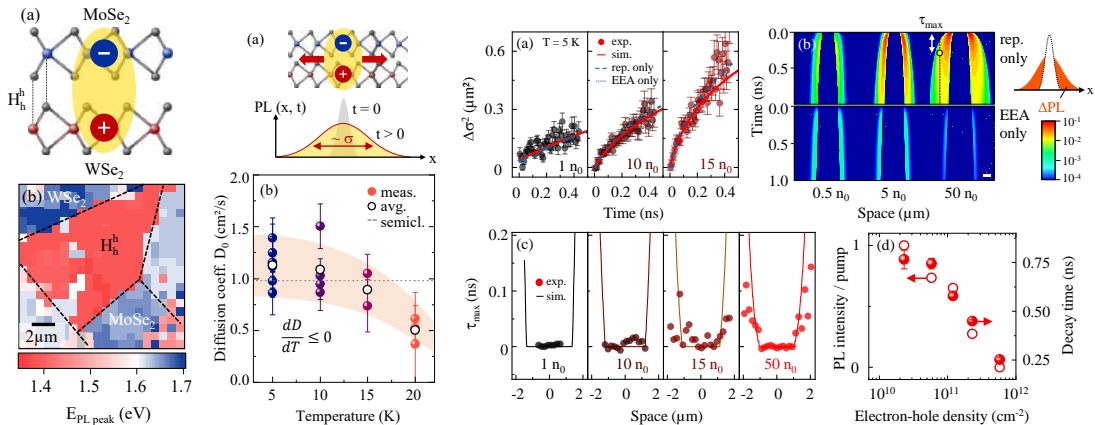
Z. Sun et al., *Nat. Photon.* 16, 79 (2022)

[A. Kis lab]

Enhancement of exciton diffusion by interactions: repulsion in BLs

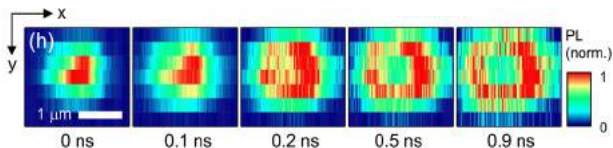
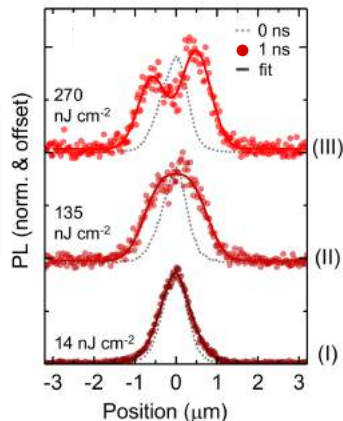
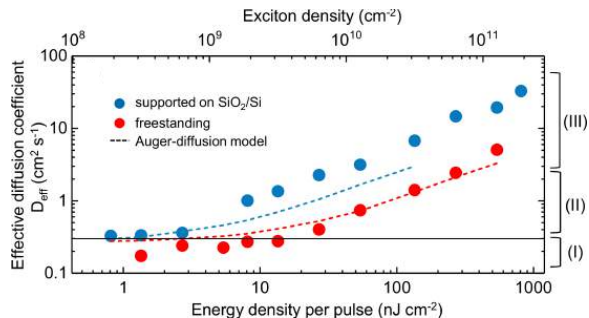
Interactions and Auger recombination

$$\frac{\partial n}{\partial t} + \frac{n}{\tau} + R_A n^2 = D \Delta n + \frac{U_0 D}{k_B T} \nabla \cdot (n \nabla n)$$



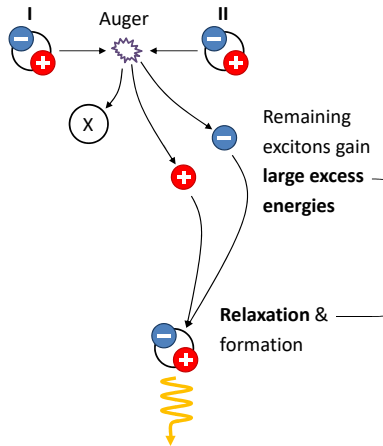
Two contributions to $D_{eff} \propto N$ are disentangled: (i) Auger effect and (ii) exciton repulsion

Halo formation at high excitonic densities as a result of Auger effect



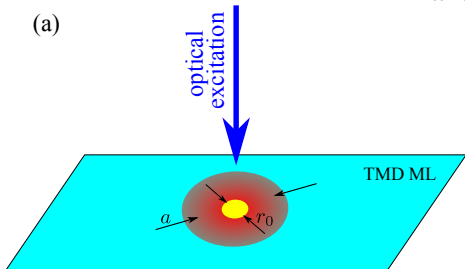
Phys. Rev. Lett. **120**, 207401 (2018); *Phys. Rev. B* **101**, 115430 (2020)

Heating via Auger scattering is the key nonlinear feedback effect



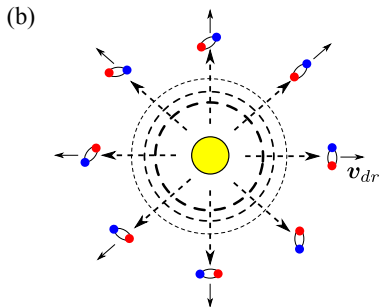
Hot spot: non-equilibrium phonons

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



- 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

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

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

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

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

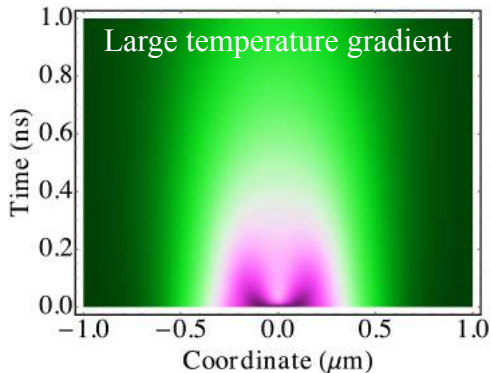
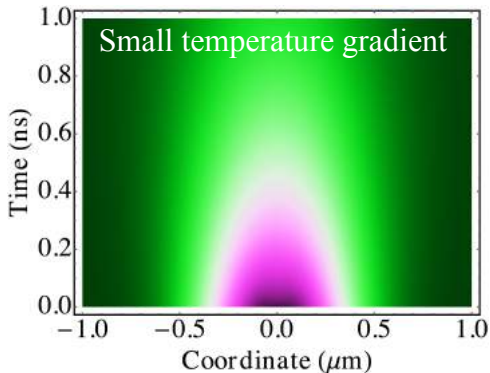
Phonon wind, drag and Seebeck effects

High temperatures, diffusive phonons

Temperature gradients of lattice and of excitons are formed

$$F_{\text{drag}} = -\frac{\tau_p}{\tau_x} k_B \nabla T_{\text{latt}}, \quad F_{\text{Seebeck}} = \frac{\mu}{k_B T} k_B \nabla T_{\text{exc}}$$

Phonon drag scenario:



MMG, *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

$$F_{\text{wind}}(\rho) = \frac{U \rho}{\rho \rho}$$



These bluestripe snapper (кашмирский луциан) are schooling (стая).

They are all swimming in the same direction in a coordinated way.

[details](#)

Phonon drag

Phonons propagate diffusively

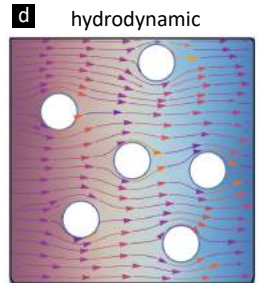
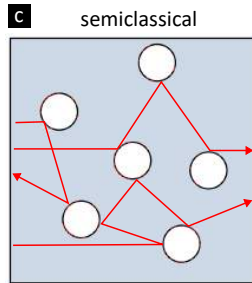
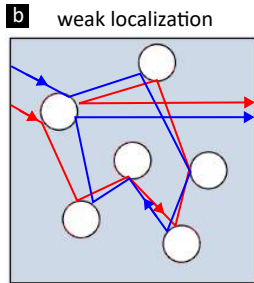
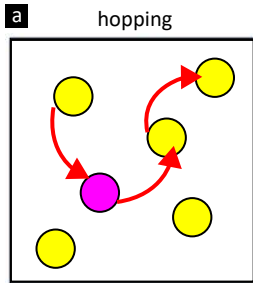
$$F_{\text{drag}}(\rho) = \nabla_{\rho} \frac{\Theta}{4\pi\chi t} \exp\left(-\frac{\rho^2}{4\chi t}\right)$$



These surgeonfish (рыба-хирург) are shoaling (скопление). They are

swimming somewhat independently, but in such a way that they stay connected.

Regimes of exciton propagation



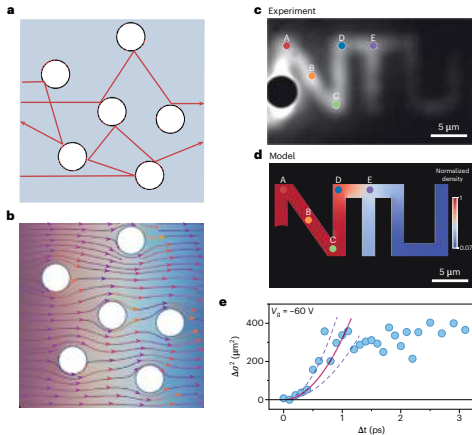
Ultrafast exciton propagation: experimental motivation

Usually exciton transport in 2D semiconductors is diffusive (large mass, efficient scattering)

Exciton diffusion in 2D van der Waals semiconductors, in 2D Excitonic Materials and Devices, ed. by P.B. Deotare and Z. Mi, Elsevier (2023).

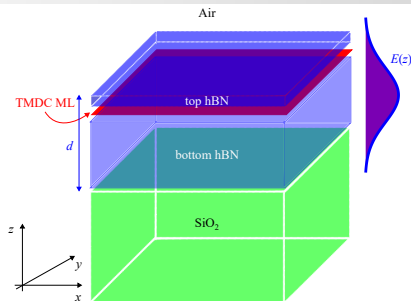
Strong exciton-exciton interactions $\tau_{xx} \ll \tau_{x-ph}, \dots \Rightarrow$ collective fluid-like behavior

del Águila, et al., Nat. Nano. (2023); MMG, Nat. Nano. (2023) + Butov's group experiments on TMDC (2021-23) + some other works



Ultimate fate at $n \gg Mk_B T / \hbar^2$ is superfluidity, but $v_{\text{exp}} \approx 0.07c$ is too high ...

Gergel, Kazarinov, Suris, JETP 27, 159 (1968); Lozovik, Yudson, JETP Letters. 22, 274 (1975); Fogler et al., Nat. Commun. 5, 4555 (2014)



Propagation process

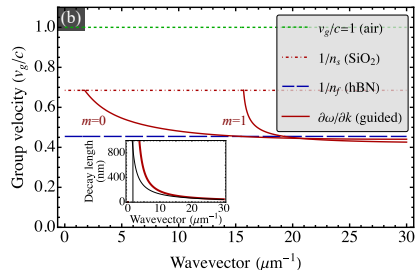
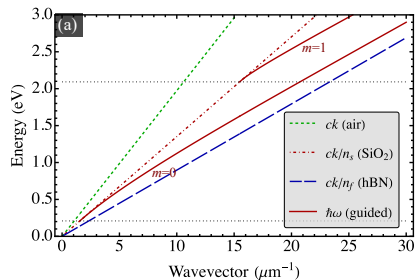
incident photon

→ scattering (roughnesses, excitons)

→ photon in a waveguide mode

→ exciton/scattering

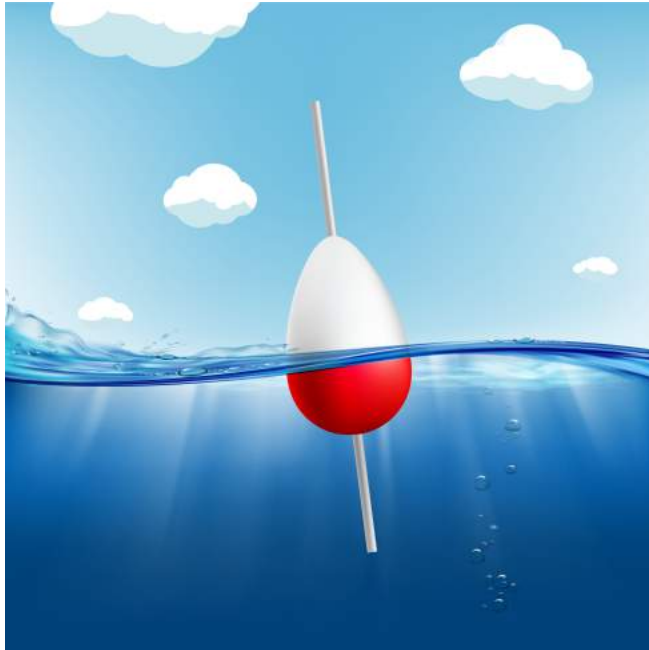
→ secondary photon



Waveguide modes with the group velocity $\sim 0.5c$ can be formed in hBN layers

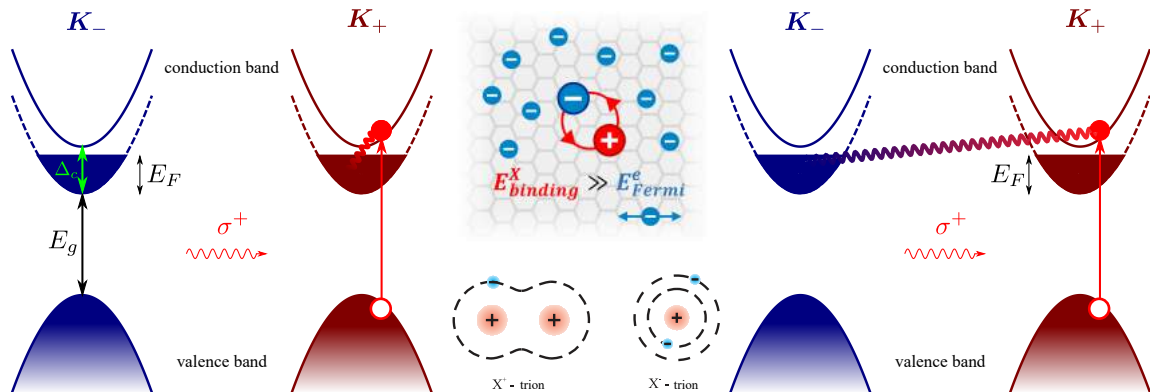
In some cases, ultrafast propagation may be related with photon transport via these modes

Exciton floating in the Fermi sea of electrons: artistic view



Bose-Fermi mixtures of excitons and carriers: Trions & Fermi polarons

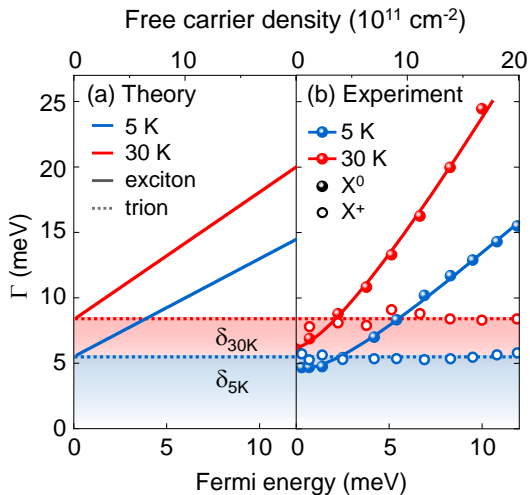
Exciton interacts with resident charge carriers



What are the diffusion mechanisms in Bose-Fermi mixtures?

details

Linewidths: experiment



Attractive polaron/trion

$$\Gamma_T = \delta$$

Repulsive polaron/exciton

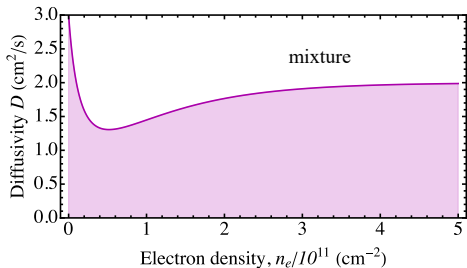
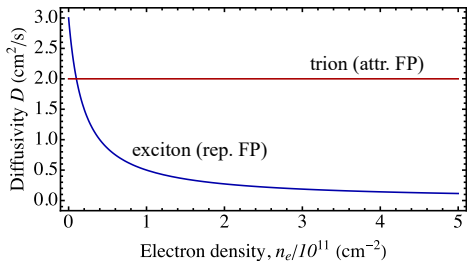
$$\Gamma_X = \delta + E_F \frac{M_T}{M_X} \frac{\pi}{\ln^2[\delta/(2E_{b,T})] + \pi^2/4}$$

Attractive polaron (trion) linewidth is practically independent of free carrier density

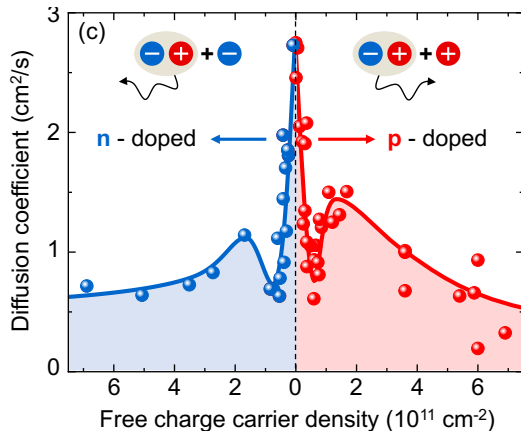
Repulsive polaron (exciton) linewidth increases with increasing the density

The increase rate depends on the temperature via $\delta(T)$

Transport in Bose-Fermi mixtures: prediction & observation

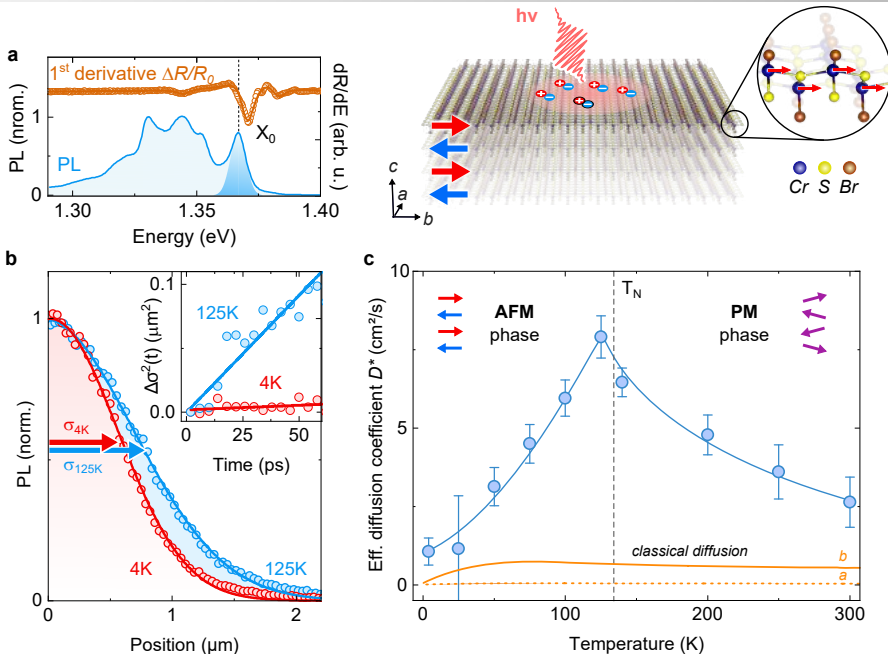


semiclassical model $D_{cl} = \frac{k_B T}{M} \tau_p$, $\tau_p = \frac{\hbar}{\Gamma}$

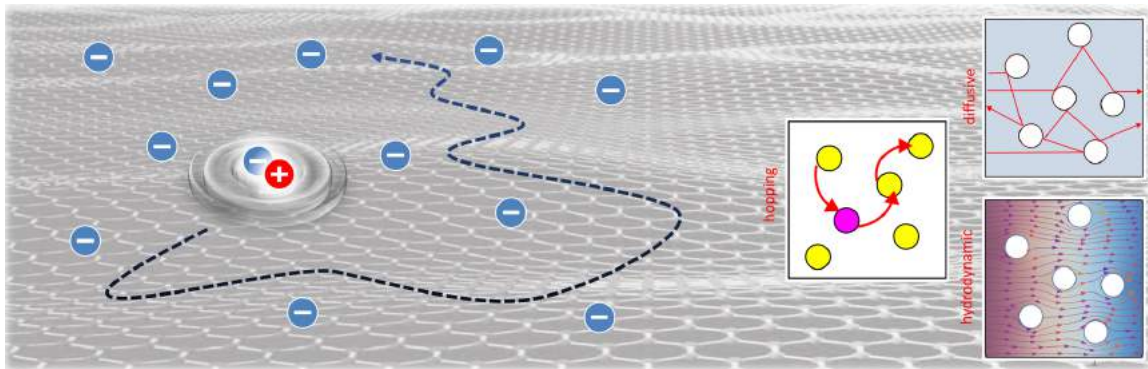


Transition between the $X - e$ scattering to the trion (FP) formation with increasing density

Exciton and magnon transport in CrSBr – van der Waals antiferromagnet



Regimes of exciton propagation



weak (free propagation) \Leftarrow **Disorder strength** \Rightarrow strong (localization, hopping)

XX-interaction strength \Rightarrow enhanced diffusivity \Rightarrow hydrodynamics (superfluidity)

XE-interaction strength \Rightarrow polaron formation

- 1 Режимы экситонного транспорта
 - Прыжковый транспорт
 - Полуклассическое распространение
 - Слабая локализация экситонов
- 2 Нелинейный транспорт экситонов
 - Экситон-экситонные столкновения
 - Экситонные жидкости
- 3 Диффузия экситонов в море Ферми электронов

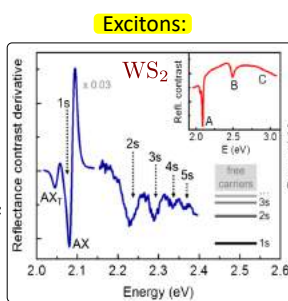
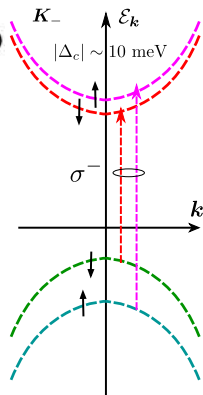
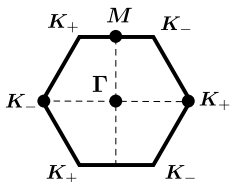
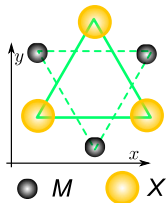
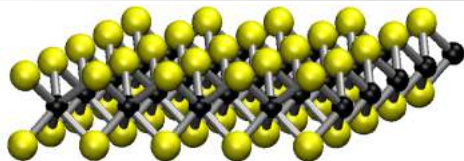


Открытые вопросы

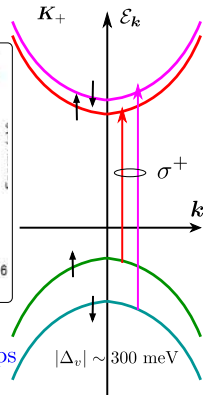
- Слабая локализация: расхождение теории и эксперимента
- Фотопроводимость: дрейф трионов/ферми-поляронов
- Гидродинамика экситонов и сверхбыстрый транспорт
- Новые системы – магнитный CrSBr:

взаимное увлечение экситонов и магнонов

Двумерные материалы – экситонные эффекты



$E_{2B} \approx 200 \dots 500 \text{ meV}$
 $a_B \approx 10 \dots 30 \text{ \AA}, \tau_X \sim 1 \text{ ps}$



- Прямозонные полупроводники $E_g \approx 2 \text{ эВ}$
- Две долины K_+ и K_-
- Спин-орбитальное взаимодействие
- Киральные оптические правила отбора
- Кулоновские эффекты: экситоны

- Платформа для ван-дер-ваальсовых гетероструктур *Geim, Grigorieva (2013)*
- Транзисторы *Radisavljevic, ..., Kis (2011)*
- Лазеры и однофотонные источники *Wu, ..., Xu (2015); Koperski, ..., Potemski (2015)*
- Сочетание необычных оптических и транспортных свойств



- **ФТИ им. А.Ф. Иоффе:** З.А. Яковлев, М.В. Дурнев, Л.Е. Голуб, М.А. Семина, Е.Л. Ивченко
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- **ФИАН им. П.Н. Лебедева:** В.В. Белых, М.В. Кочиев
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- **Phillips-Marburg Uni./U. Chalmers:** R. Rosati, R. Perea-Causín, E. Malic
- **INSA-Toulouse:** G. Wang, S. Shree, C. Robert, X. Marie, B. Urbaszek, T. Amand
- **IOP Beijing:** C.R. Zhu, B. Liu
- **Weizmann Institute of Science:** Sivan Refaely-Abramson



Экситоны в двумерных материалах

М.М. Глазов

ФТИ им. А.Ф. Иоффе, Санкт-Петербург

- 1 Двумерные диалкогениды переходных металлов
- 2 Теория экситонов Ванье-Мотта
- 3 Особенности кулоновского взаимодействия и экситонной серии в 2D
- 4 Тонкая структура экситонных состояний
- 5 Взаимодействие экситонов и электронов: трионы и ферми-поляроны
- 6 Пара слов о том, как экситоны взаимодействуют друг с другом
- 7 Экситоны, фононы и упругие деформации
- 8 Экситонный транспорт: классические и квантовые эффекты

За кадром:

гетероструктуры, муар, коррелированные электронные и экситонные фазы, магнетизм

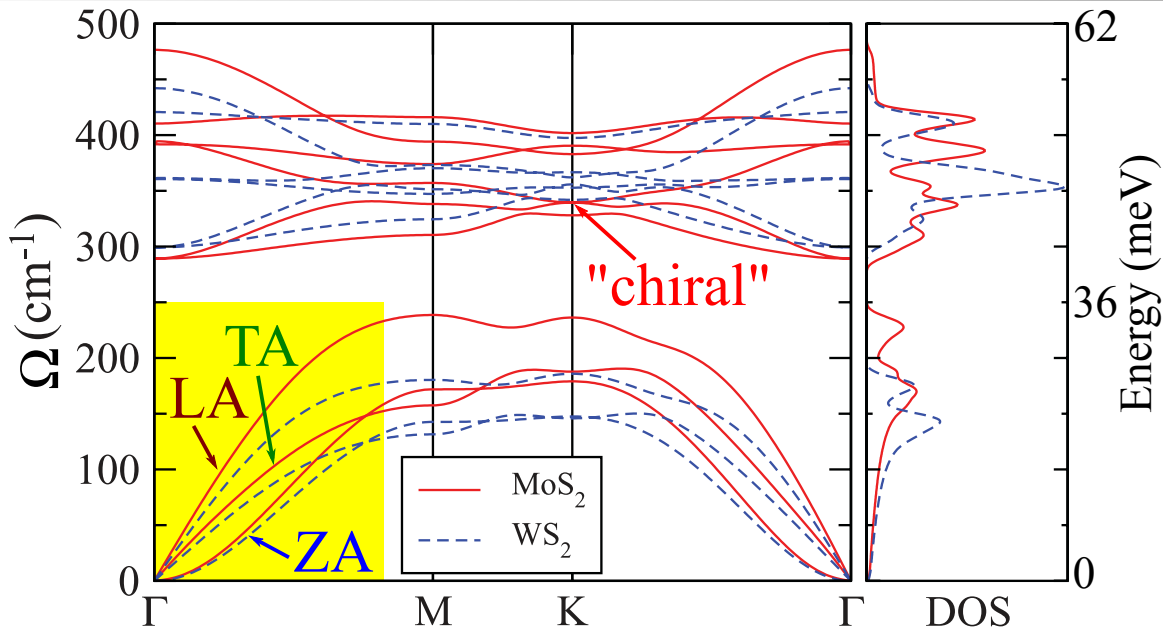
**ЛЕТНЯЯ ШКОЛА
ФОНДА «БАЗИС»**

БАЗИС

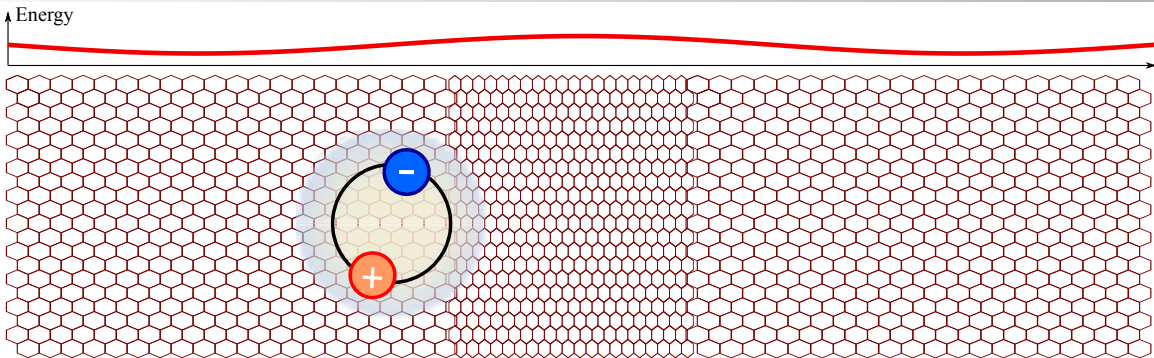


Российский фонд
научных исследований
197-1000-1000-1000

Vibration spectrum



Deformation potential



Lattice deformation \Rightarrow shift of the electron and hole energies \Rightarrow variation of exciton energy

$$\Delta E_x = (\Xi_c - \Xi_v)(\epsilon_{xx} + \epsilon_{yy}) \propto (\mathbf{q} \cdot \mathbf{u}_q)$$

Matrix element:

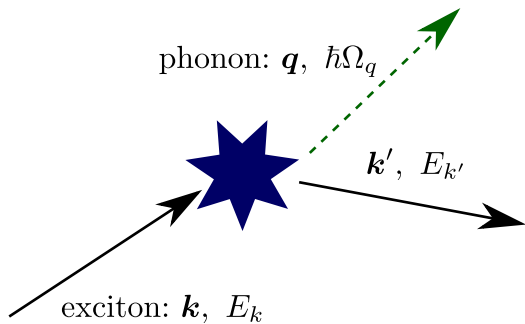
$$M_{k'k}^q = \sqrt{\frac{\hbar}{2\rho\Omega_q S}} q(D_c - D_v)\mathcal{F}(q), \quad \mathcal{F}(q) \approx \frac{1}{[1 + (qa_B/4)^2]^{3/2}} \approx 1$$

effective for LA mode; piezo interaction [$\propto \sqrt{q}/(1 + r_0q)$ in 2D] is weak

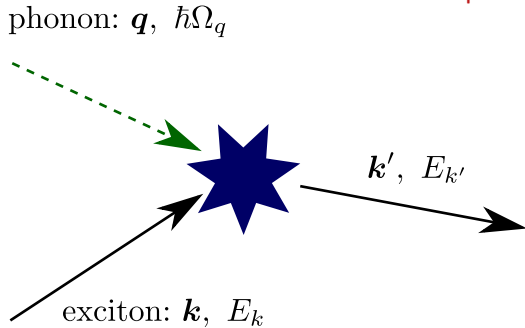
Exciton-phonon scattering rates

Scattering rates

Emission



Absorption



Quasi-elastic scattering

at $k_B T \gg M s^2 \sim 100 \mu\text{eV}$ the involved phonon energy $\hbar\Omega_{\mathbf{q}} \ll k_B T$
(M is the exciton mass, s is the speed of sound)

$$\frac{1}{\tau_p} = \frac{2\pi}{\hbar} \sum_{\mathbf{q}} |M_{\mathbf{k}'\mathbf{k}}^{\mathbf{q}}|^2 (1 - \cos\theta) (1 + 2n_{\mathbf{q}}) \delta(E_{\mathbf{k}} - E_{\mathbf{k}'}) = c \frac{k_B T}{\hbar} \quad (c \sim 1)$$

Energy relaxation rate: $\frac{1}{\tau_{\epsilon}} = \frac{2M^2(\Xi_c - \Xi_v)^2}{\hbar^3 \rho} \sim \frac{1}{10 \text{ ps}} \ll \frac{1}{\tau_p} \quad @ T \gtrsim 2 \text{ K}$

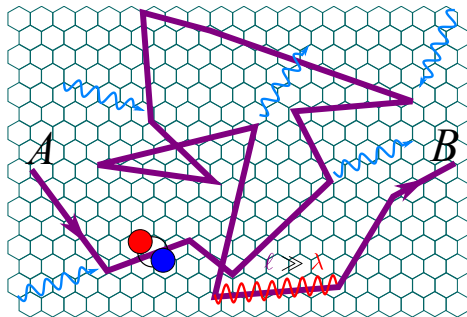
Semiclassics for TMDCs in a nutshell

$$D = \int_0^\infty \langle \hat{v}_x(t) \hat{v}_x(0) \rangle dt = \left\langle \frac{v^2 \tau_p}{2} \right\rangle$$

$$\text{semiclassics: } \ell \gg \lambda \Leftrightarrow \frac{k_B T \tau_p}{\hbar} \gg 1$$

diffusion coefficient from velocity correlator

$$\langle v_x(t) v_x(0) \rangle = v_x^2(0) e^{-t/\tau_p}, \quad \frac{1}{\tau_p} = \sum_{k'} W_{kk'} (1 - \cos \vartheta)$$



LA-phonon scattering in MX_2 MLs

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

Here D is temperature independent; **experiment at 4 K gives similar values ($\sim 2.5 \text{ cm}^2/\text{s}$).**

Time scales

Quasielasticity:

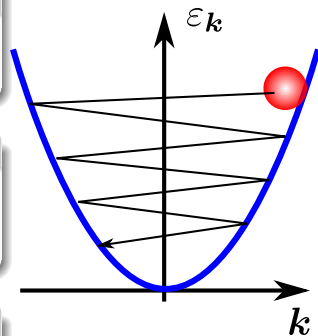
$$\Delta\epsilon \sim \sqrt{k_B T M s^2} \ll k_B T \Rightarrow \delta\epsilon^2(t) \sim (\Delta\epsilon)^2 \frac{t}{\tau}$$

Momentum relaxation time

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

Energy relaxation time

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



$$\tau_\epsilon \gg \tau_\phi \gg \tau$$

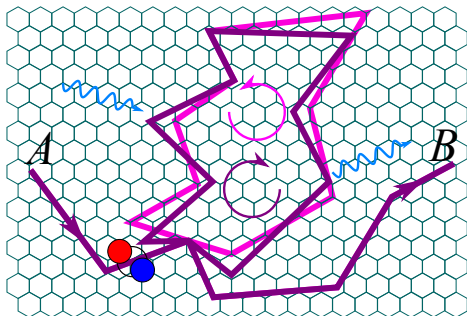
Phase relaxation time

Altshuler, Aronov, Khmel'nitsky (1981)

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

Acoustic phonon scattering: quantum effects get stronger with the temperature increase

Exciton weak localization: Dephasing



$$D = \hat{v}_x \text{---} \hat{v}_x$$

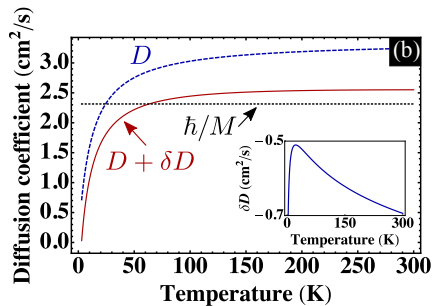
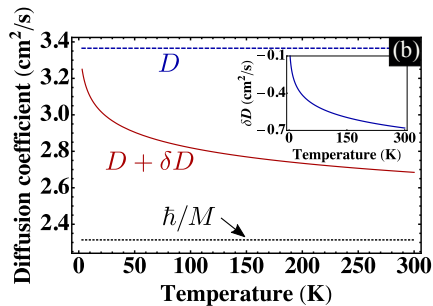
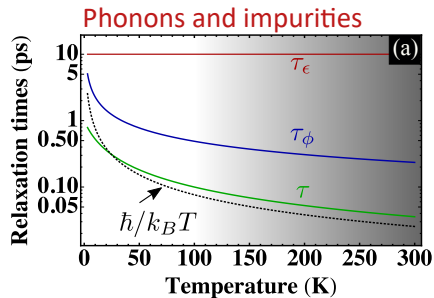
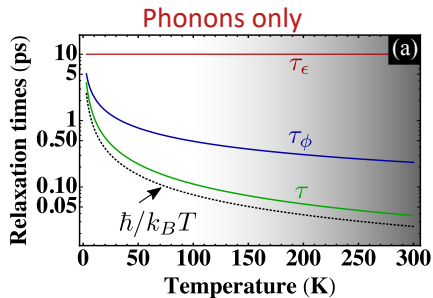
$$\delta D = \hat{v}_x \text{---} \text{X} \text{---} \hat{v}_x$$

Interference amplitude (Cooperon):

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

$$\frac{\delta D}{D} \sim -\frac{\hbar}{k_B T \tau} \ln \left(\frac{\tau_\phi}{\tau} \right), \quad \frac{\hbar}{\tau_\phi} \sim \delta\epsilon(\tau_\phi), \quad \frac{\tau_\phi}{\tau} = \sqrt[3]{\frac{\hbar^2}{Ms^2\tau^2 k_B T}} \propto T^{1/3}$$

Weak localization: results



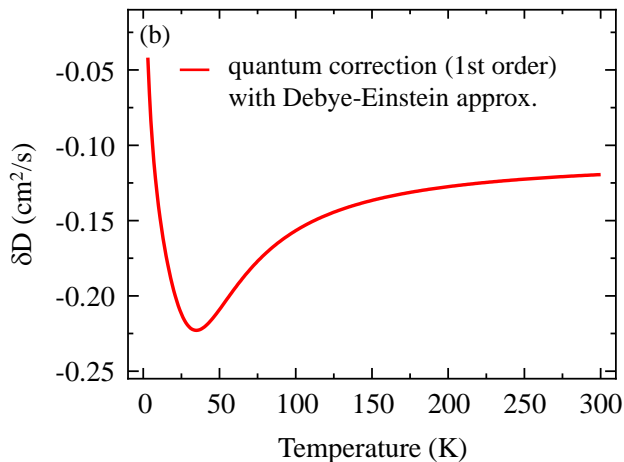
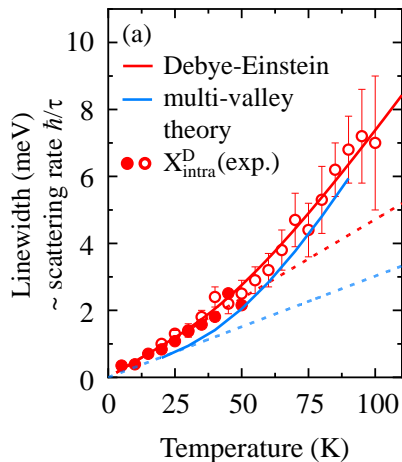
PRL 124, 166802 (2020)

Various scattering mechanisms

Scattering mechanism	$D_{cl}(T)$	τ_ϕ/τ	$\delta D(T) < 0$
LA	T^0	$T^{1/3}$	↓
LA + disorder	T	$T^{-2/3}$	↑
flexural, 1ML	T^0	T^0	const
flexural, 2ML, $T < T_0$	T^0	$T^{2/3}$	↓
flexural, 2ML, $T > T_0$	$T^{3/2}$	$T^{-5/6}$	↑
LA, overdamped	T^0	$T^{1/2}$	↓
LA + free e/h	T^0	T/n	↓

Appl. Phys. Lett. **121**, 192106 (2022)

Experiment: non-classical exciton propagation – II

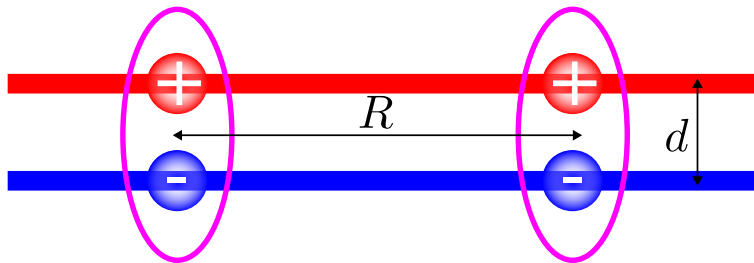


PRL 127, 076801 (2021)

Dipolar repulsion of excitons in bilayers

Direct Coulomb interaction

$$U(R) \approx \frac{2e^2}{\epsilon} \left(\frac{1}{R} - \frac{1}{\sqrt{R^2 + d^2}} \right), \quad V = \int U(R) d^2R = \frac{4\pi e^2 d}{\epsilon}$$



Interaction-induced blueshift $\Delta E(n) = Vn$ “plate capacitor model”

Butov, Shashkin, Dolgoplov, Campman, Gossard (1999)

Exciton-exciton correlations: $V \rightarrow \mathcal{K}(T)V$, $\mathcal{K}(T) \approx 0.1 \dots 1$

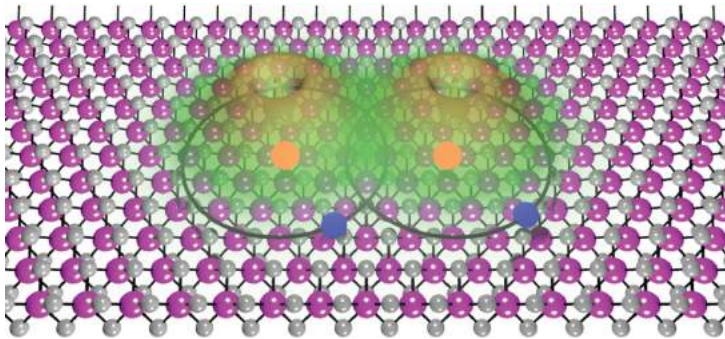
Zimmermann, Schindler (2007,2008); Laikhtman, Rapaport (2009)

+ band gap renormalization and screening ...

2D semiconductors: *Erkensten, Brem, Perea-Causin, Malic (2022)*

Exchange interaction of excitons in monolayers

In monolayers direct Coulomb interaction is suppressed due to the charge neutrality



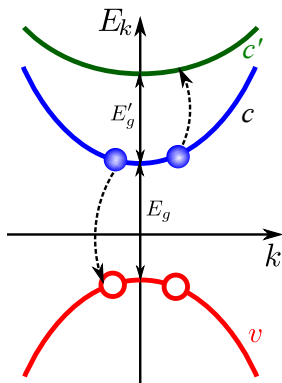
Exchange contribution \Rightarrow overlap of the wavefunctions:

$$V_{\uparrow\uparrow} \sim E_B a_B^2, \quad V_{\uparrow\downarrow} \sim \frac{\hbar^2}{M} \ln \left(-\frac{E}{E_{bi}} \right)$$

Quantum wells: Ciuti, Savona, Piermarocchi, Quattropani, Schwendimann (1998); Combescot, Betbeder-Matibet, Dubin (2008);
Microcavities: Tassone, Yamamoto (1999); MMG, Ouerdane, Pilozi, Malpuech, Kavokin, D'Andrea (2009);
2D semiconductors: Shahnazaryan, Iorsh, Shelykh, Kyriienko (2017)

Exciton-exciton annihilation: Auger-like process

Resonant interaction of excitons



Conservation laws:

$$E_1 + E_2 = E_f, \quad \mathbf{K}_1 + \mathbf{K}_2 = \mathbf{K}_f$$

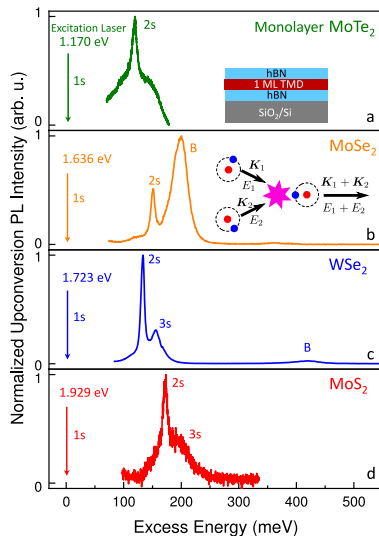
Resonance: $E'_g \approx E_g - E_B$

$$\frac{dn_{cv}}{dt} = -R_A n_{cv}^2 = -\frac{dn_{c'v}}{dt}$$

$$R_A \propto \frac{E_B^2}{k_B T} \left| \frac{p_{cv} p_{c'v}}{E_g E_{g'}} \right|^2 e^{-|\delta|/k_B T}$$

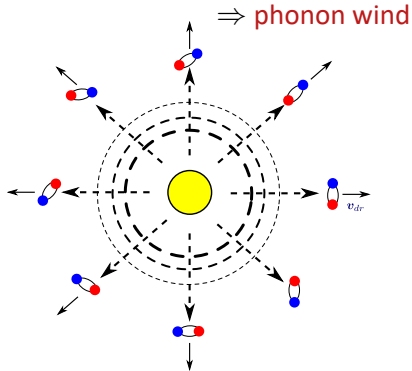
Resonant Auger process populates excited states
(photoluminescence upconversion)

(eV)	MoS ₂	MoSe ₂	WSe ₂	MoTe ₂
E_g	1.8	1.6	1.7	1.7
E'_g	1.2	1	1.4	1.3
E_B	0.2	0.18	0.16	0.16



Low temperatures, ballistic phonons

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



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 $N_q \gg 1$

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

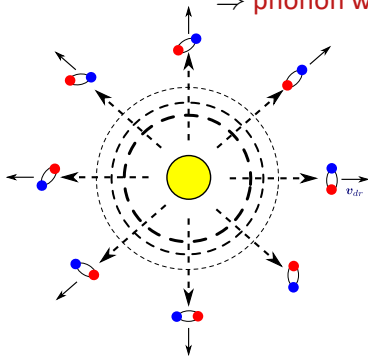
$$[N_q \delta(E_{k+q} - E_k - \hbar\Omega_q) + N_{-q} \delta(E_{k+q} - E_k + \hbar\Omega_q)]$$

Phonon wind effect

Low temperatures, ballistic phonons

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

⇒ phonon wind



Phonons drag excitons away

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

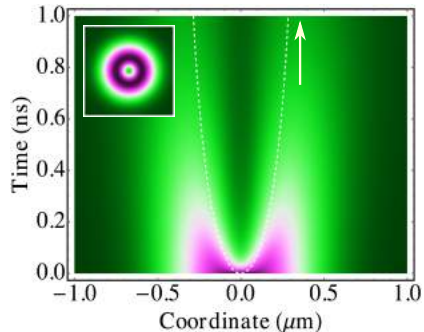
Force field produced by phonons (2D)

$$F_{\text{wind}}(\rho) = \frac{U \rho}{\rho \rho}$$

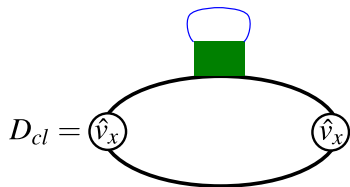
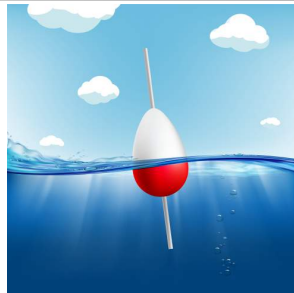
Hot spot acts as a repulsive center

effective “Coulomb” repulsion

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



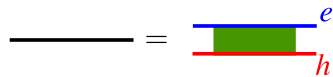
$$D = \int_0^\infty \langle\langle \hat{v}_x(t) \hat{v}_x(0) \rangle\rangle dt, \quad \langle v_x(t) v_x(0) \rangle = v_x^2 e^{-|t|/\tau}$$



Semiclassical approach (short-range scattering)

$$D = \left\langle \frac{v^2 \tau}{2} \right\rangle, \quad \frac{1}{\tau} = \frac{2}{\hbar} \text{Im} \Sigma(E_k^x, \mathbf{k})$$

Scattering contribution to the linewidth



$$\Gamma = \frac{2}{\hbar} \text{Im} \Sigma(0, 0) \left(= \frac{1}{\tau} \text{ for many scattering mechanisms} \right)$$

low temperatures, phonons are suppressed: τ is the exciton-electron scattering time

Trion/Fermi polaron scattering

Chevy ansatz:

$$\Psi_{\mathbf{k}} = \varphi(\mathbf{k}) X_{\mathbf{k}}^{\dagger} |0\rangle + \sum_{p,q} \underbrace{F_{p,q}(\mathbf{k}) X_{\mathbf{k}+q-p}^{\dagger}}_{\text{trion}} \underbrace{e_p^{\dagger}}_{\text{FS-hole}} e_q |0\rangle$$

Short-range interaction model:

$$\varphi(\mathbf{k}) \approx \frac{V_0 N_e}{-E_{b,tr}} \mathcal{F}(\mathbf{k}), \quad F_{p,q}(\mathbf{k}) \approx \frac{V_0}{E_{\mathbf{k}}^{tr} - E_{\mathbf{k}-p}^x - E_p^e} \mathcal{F}(\mathbf{k}), \quad |\mathcal{F}(\mathbf{k})|^2 \approx \frac{E_{b,tr}}{N_e \mathcal{D} V_0^2}.$$

Perturbation (external field)

$$\hat{V} = \sum_{\mathbf{k},\mathbf{k}'} V_x(\mathbf{k}' - \mathbf{k}) X_{\mathbf{k}'}^{\dagger} X_{\mathbf{k}} + \sum_{\mathbf{p}',\mathbf{p}} V_e(\mathbf{p}' - \mathbf{p}) e_{\mathbf{p}'}^{\dagger} e_{\mathbf{p}}; \quad V_x = V_e + V_h$$

Large momentum transfer $|\mathbf{k} - \mathbf{k}'| \gg k_F$

$$V_{\mathbf{k},\mathbf{k}'} \approx V_x + V_e$$

Small momentum transfer $|\mathbf{k} - \mathbf{k}'| \leq k_F$

$$V_{\mathbf{k},\mathbf{k}'} \approx V_x + V_e - V_{FS-h} \approx V_x.$$

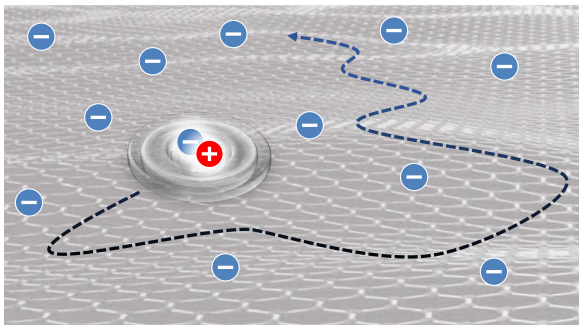
Repulsive (exciton) and attractive (trion) polaron scattering

Related problem: lifetimes of polaron quasiparticles *Petrov (2003); Yan (2019); Cotlet et al. (2019); Adlong (2020)*

At low temperatures, the main scattering is due to the resident electrons

$$\text{Im } T(\varepsilon, \mathbf{k}) = \left| \frac{V_0}{1 - V_0 S(\varepsilon, \mathbf{k})} \right|^2 \text{Im } S(\varepsilon, \mathbf{k})$$

$$\text{Im } S(\varepsilon, \mathbf{k}) = \pi \sum_{p'} (1 - n_{p'}) \delta(\varepsilon - E_{p'}^e - E_{\mathbf{k}-p'}^x) = \begin{cases} \pi D_{\text{eff}}, & \varepsilon > 0 \text{ repulsive polaron/exciton} \\ 0, & \varepsilon < 0 \text{ attractive polaron/trion} \end{cases}$$



For repulsive polaron/exciton

$$\text{Im } \Sigma(\varepsilon > 0) \neq 0 \Rightarrow$$

effective scattering by electrons

For attractive polaron/trion

$$\text{Im } \Sigma(\varepsilon < 0) = 0 \Rightarrow$$

only higher-order (weaker) processes
and phonon + disorder scattering

Electron-exciton scattering

Clean limit (Fermi's golden rule):

$$\text{Im } \Sigma_x(E_k^x, \mathbf{k}) = \pi \sum_{p, p'} n_p (1 - n_{p'}) \delta(E_k^x + E_p^e - E_{p'}^e - E_{k+p-p'}^x) \left| \frac{V_0}{1 - V_0 S(\varepsilon, \mathbf{k})} \right|^2 \Rightarrow$$
$$\Gamma_x(k) \propto \frac{(k_B T)^{3/2}}{\sqrt{N_e}} \rightarrow 0 @ k_B T \ll E_F \quad (\text{Fermi liquid-like behavior})$$

Pauli blocking prevents exciton-electron scattering

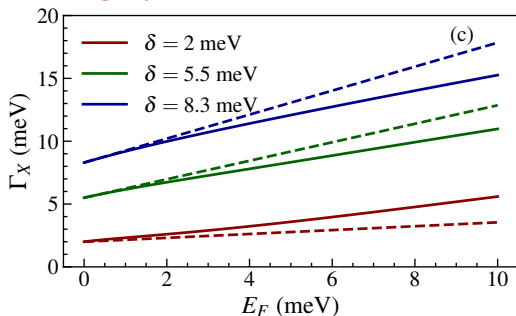
cf. Cotleț et al. (2019); higher-order processes: Petrov (2003); Adlong (2020)

Dirty limit, $\delta \gg k_B T$ (acoustic phonon or impurity scattering is present)

$$T(\varepsilon, 0) \approx \frac{1}{D} \frac{1}{\ln\left(-\frac{E_{b,T}}{\varepsilon + i\delta/2}\right)}$$

$$\Gamma_X = \delta + E_F \frac{M_T}{M_X} \frac{\pi}{\ln^2[\delta / (2E_{b,T})] + \pi^2 / 4}$$

numerics: Efimkin & MacDonald (2017); Rana et al. (2020); Katsch & Knorr (2022)



Additional scattering breaks energy-momentum conservation
and enhances electron-exciton scattering