Excitation-induced dephasing in 2D materials and van der Waals heterostructures

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Excitons dominate the excitation kinetics in transition-metal dichalcogenides (TMDs). As co-bosonic particles consisting of electrons and holes, excitons show a non-trivial scattering dynamics. Here, we study exciton-exciton interaction and excitation induced dephasing (EID) on microscopic footing and predict efficient scattering for momentum-dark excitons because of a large electron/hole mass asymmetry. We find an unexpected temperature dependence of the EID and an orders of magnitude stronger interaction of interlayer excitons in TMD heterostructures due to their permanent dipole moment.

The emergence of atomically thin 2D materials, such as graphene and monolayer transition metal dichalcogenides (TMDs), has initiated a new research field offering a platform for the investigation of intriguing many-body correlations and quantum phenomena [1–4]. The strong Coulomb interaction in atomically thin TMD monolayers gives rise to tightly bound excitons dominating the optical response, relaxation dynamics and transport characteristics [5–8]. Excitons are neutral composite bosons (cobosons) consisting of Coulomb-bound electrons and holes. Due to a complex band structure exhibiting multiple valleys in TMDs, there is a variety of different exciton species including bright, momentum- and spin-dark intralayer excitons [9–11] as well as spatially separated interlayer excitons in van der Waals heterostructures [12–15].

Previous theoretical studies have addressed the fundamental many-body processes governing the phonon-driven exciton dynamics in monolayer TMDs [11, 16–19] and van der Waals heterostructures [20–22]. In the considered weak-excitation regime, excitons were treated as non-interacting bosons. However, as the excitation density increases, the cobosonic nature of excitons comes to the surface and exciton-exciton scattering becomes increasingly important [23–25]. While scattering between electrons has been treated extensively in literature [26, 27], a microscopic treatment of the Coulomb interaction of excitons as neutral cobosonic quasi-particles has proven to be highly challenging [28–30]. In particular, exciton-exciton scattering incorporating the remarkable intervalley and interlayer excitonic landscape in 2D materials has remained unrevealed.

In this Letter, we investigate exciton-exciton scattering in TMD monolayers and van der Waals heterostructures based on a microscopic and fully quantum-mechanic approach. We calculate the excitation-induced dephasing (EID) resolving the underlying intra- and intervalley as well as intra- and interlayer exciton-exciton scattering channels, cf. Fig. 1. We reveal an intriguing temperature and screening dependence of EID and provide microscopic insights into the fundamental nature of scattering between intra- and interlayer excitons. The latter exhibit a permanent out-of-plane dipole moment and their interaction can be considered as an efficient dipole-dipole coupling (Fig. 1(a)) resulting in an EID in the range of a few meV. In contrast, intralayer excitons do not have a permanent dipole moment and interact through higher-order electric moments induced by the internal charge inhomogeneity of excitons as cobosonic quasi-particles. Here, we show that the mass asymmetry between electron and holes as well as the overlap of excitonic Bohr radii are the key quantities determining the exciton-exciton scattering efficiency. As a direct consequence, we predict most efficient scattering with momentum-dark intervalley excitons exhibiting a large electron/hole mass asymmetry. The gained microscopic insights are applicable to a broader class of excitonic, multi-valley materials.

Model.— To be able to investigate exciton-exciton scattering on a microscopic footing in 2D materials, we first define the many-particle Hamilton operator. Following the approach described in Ref. 30, we derive the excitonic Hamilton operator from the conventional electron-electron Hamiltonian by employing an identity operator expansion [31] and rewriting the electron and hole creation and annihilation operators $c^{(\dagger)}$ and $\psi^{(\dagger)}$ into excitonic operators $P^{(\dagger)}$. We obtain $H = H_0 + H_{\alpha-\beta}$.
where the interaction-free part $H_0$ includes the excitonic eigenenergies $\epsilon_Q$ with $\alpha = \{n, \xi\}$. The latter is a compound index including the excitonic state $n = 1s, 2s...$ in the valley $\xi = (\xi_h, \xi_e) = \text{KK}, \text{KK}', \text{KA}$. The eigenenergies and the associated excitonic wave functions $\phi_{\alpha, q}$ are obtained by solving the Wannier equation [16, 20, 32]. In the case of monolayers, we treat the screened Coulomb potential entering the Wannier equation within the Keldysh approach [33–35] to account for the finite width of the TMD and screening effects. For charge carriers within a heterostructure, we solve the Poisson equation for two aligned homogeneous slabs resulting in a generalized Keldysh screening [20].

The exciton-exciton interaction $H_{x-x}$ is given by

$$H_{x-x} = \frac{1}{2} \sum_{q} V^{\alpha\beta\alpha'\beta'}_{q} P_{\alpha, Q+q}^\dagger P_{\beta', Q-q}^\dagger P_{\beta, Q'-q} P_{\alpha', Q'},$$

where summation over the excitonic indices $\alpha^{(0)}, \beta^{(0)}$ and momenta $Q^{(0)}, q$ is implied [30]. The expression includes direct electron-electron, hole-hole, and electron-hole interactions summarized in the excitonic Coulomb matrix element $V^{\alpha\beta\alpha'\beta'}_{q}$. In this work, we focus on the most relevant valley-conserving exciton-exciton scattering processes ($\alpha = \alpha' \equiv \mu$ and $\beta = \beta' \equiv \nu$) involving a small momentum transfer in the energetically lowest $n = 1s$ state. Figure 1(b) illustrates exemplary scattering processes involving only KK states (process I) and including different valleys (process II). The corresponding monolayer exciton-exciton Coulomb matrix element reads

$$V^{\mu\nu\mu\nu}_{q}|_{\text{mono}} = W_{q} D_{\mu}(q) D_{\nu}^*(q),$$

with the screened Coulomb potential $W_{q}$ and $D_{\mu}(q) = (F_{\mu}(\beta^0 q) - F_{\mu}^*(\alpha^0 q))$ including the form factors

$$F_{\mu}(x q) = \sum_{q} \phi_{\mu, q}^{\dagger} e^{i q \cdot r} \phi_{\mu, q}.$$

The latter describe the overlap of excitonic wave functions including the mass ratios $\alpha = m_{e(h)}^{\nu}/m_{e(h)}^{\mu}$, $\beta = 1 - \alpha$, where $m_{e(h)}^{\mu}$ is the electron (hole) mass in the valley $\mu = K, \Lambda, K'$. When considering scattering processes within a heterostructure the form factors are weighted differently depending on whether the scattering occurs between interlayer excitons exclusively or between intralayer and interlayer excitons, cf. the Supplemental Material.

Figure 2(a)-(b) illustrates the exciton-exciton Coulomb matrix element $V^{\mu\nu\mu\nu}$ in a hBN-encapsulated WSe$_2$ monolayer considering different scattering channels with $\mu = \text{KK}$ and $\nu = \text{KK}, \text{KA}, \text{KK}'$ in real and momentum space. Since excitons are neutral quasi-particles composed by electrons and holes, the resulting interaction potential in real space is reminiscent of the Lennard-Jones potential [36]. We find that both repulsive or attractive exciton-exciton interactions occur for different exciton species and length scales. Our model predicts that the strength of the interaction is determined by the mass ratios of electrons and holes of the involved excitons. This can be seen by performing a Taylor expansion of Eq. (2) for small $q$ giving $V^{\mu\nu\mu\nu}_{q} \approx W_{q} r_{\mu, B}^2 r_{\nu, B}^2 q^2 Q_{\mu} Q_{\nu}$, with $r_{\mu, B}^2 = |\mu|^2 |\nu|$, where $r_{\mu, B}$ is the excitonic Bohr radius. Furthermore, we have introduced the effective excitonic charge

$$Q_{\mu} = (Q_{h} m_{h}^{\mu} + Q_{e} m_{e}^{\mu}) / (m_{h}^{\mu} + m_{e}^{\mu})$$

with $Q_{h/e} = \pm 1$ determining the sign of the interexcitonic potential and thus dictating the repulsive or attractive nature of the interaction. The latter can be interpreted as a weak force resulting from the internal charge inhomogeneity of the exciton. In particular, an exciton with a heavy hole will be positively charged in its center, surrounded by a negatively charged shell resulting from the orbiting electron. Considering the effective excitonic charges, the matrix element in the long range limit is always positive for $\mu = \nu$, while it becomes negative if the interacting excitons have inverted mass ratios. For example, we find an attractive character at small distances for the KK-KA and KK-KK’ interaction in Fig. 2(b). While holes are heavier than electrons for KK excitons ($Q_{\mu} > 0$), the opposite is the case for KA and KK’ ($Q_{\nu} < 0$). Moreover, having the hole at the K point and the electron at the $\Lambda$ point rather than at the K’-point increases the exciton-exciton interaction by a factor of 10 in momentum space, cf. Fig. 2(a). This is a direct consequence of a larger mass asymmetry and consequently larger effective charge for KA excitons ($m_{k}^K = 0.36 m_0$) and $m_{e}^K \approx 0.6 m_0$, vs $m_{e}^{K'} = 0.4 m_0$ [37]). Moreover, the interaction strongly depends on the excitonic Bohr radius (expected distance between electron and hole) as it
directly scales with the quadrupole moments of the excitons. Similarly to the monolayer case, we perform an analysis of the interlayer excitonic Coulomb matrix element in a van der Waals heterostructure. In contrast to intralayer excitons, here the repulsion between electrons/holes is stronger than the counteracting attraction of electrons and holes of different excitons. Therefore, the interaction between interlayer excitons can be understood as repulsion between two aligned electric dipoles, cf. Fig. 1(a). As a consequence, the exciton-exciton interaction between interlayer excitons in the considered exemplary MoSe$_2$-WSe$_2$ heterostructure is up to two orders of magnitude larger than in the monolayer WSe$_2$ case, cf. Fig. 2(c)-(d). Importantly, the interlayer and intralayer matrix elements also differ qualitatively. By expanding the elements for small momenta, we find that $V_{q}^{\mu\nu\mu'\nu'}_{\text{mono}} \propto \nu_{q}q^4$ vanishes for $q \to 0$, whereas the interlayer element remains non-zero, cf. Figs. 2(a) and (c) as well as the Supplemental Material for more details.

Based on the derived excitonic Coulomb matrix elements, we now calculate the depoling of optical polarisations induced by exciton-exciton scattering that is referred to in literature as excitation-induced dephasing (EID). EID is a directly accessible phenomenon in experiments and manifests itself as a density-dependent broadening of excitonic transitions. We obtain microscopic access to the EID due to exciton-exciton interactions by evaluating the Heisenberg equation of motion for the exciton polarisation $p_{\mu} = \langle \hat{P}_{\mu, Q=0} \rangle$ in the light cone, i.e. $Q = 0$. When commuting the exciton-exciton Hamiltonian $H_{x-x}$ with the polarisation, exciton-exciton correlations such as $S \propto \langle P^\dagger P \rangle$ need to be considered, which previously have been treated within the Hartree-Fock approximation [30]. Here, we go beyond and evaluate the equations of motion in second-order Born-Markov approximation [32, 38], cf. the Supplemental Material for more details. To linear order in exciton density, we obtain $p_{\mu} \sim H_{x-x} = \gamma_{Q=0}^{\mu} p_{\mu}$ introducing the excitation-induced dephasing

$$\gamma_{Q}^{\mu}(T) = \frac{\pi}{\hbar} \sum_{\nu Q q} |V_{q}^{\mu\nu\mu'\nu'}|^2 N_{Q}^{\nu}(T) \delta(\Delta \epsilon) \tag{4}$$

with the delta-function $\Delta \epsilon = \epsilon_{Q'} + \epsilon_{Q} - \epsilon_{Q' + q} + \epsilon_{Q + q}$ ensuring the energy conservation for the considered exciton-exciton scattering processes. The appearing temperature-dependent exciton occupation $N_{Q}^{\nu}(T)$ is estimated with an equilibrium Boltzmann distribution, parameterized by the total exciton density $n = \sum_{\nu Q} N_{Q}^{\nu}$.

In the following we focus on the EID of bright KK excitons and evaluate Eq. (4) for the state in the light cone, i.e. $Q = 0$. In general, the expression has to be evaluated numerically, but in the particular case, where intravalley scattering ($\mu = \nu = \text{KK}$) is dominant, the temperature and density dependence of EID can be addressed even analytically yielding

$$\gamma_{Q=0}^{\mu}(T) \big|_{\nu=\text{KK}} = \frac{n}{\hbar} \sqrt{\frac{M_{\text{KK}}}{8\pi k_B T}} \int \nu V_{q}^{\text{KK}, \mu} \nu q^2 \, dq \tag{5}$$

We see immediately a linear dependence of EID on exciton density $n$ as well as an explicit temperature dependence scaling with $T^{-\frac{1}{2}}$. Normally, temperature dependencies observed in linewidth experiments are attributed to the interaction with phonons [23]. However, our microscopic model predicts that the temperature dependent distribution of excitons in momentum space has a direct consequence on the amount of channels available for an energy and momentum conserving exciton-exciton scattering process. This results in an additional temperature dependent broadening which can be experimentally separated from the phonon-broadening by varying both density and temperature.

**Excitation-induced dephasing in monolayers.**—We exploit Eqs. (2) and (4) to determine the excitation-induced dephasing of bright KK excitons in a hBN-encapsulated WSe$_2$ monolayer. The choice of substrate is motivated by the fact that Auger processes [39–41], not captured by our theoretical model, are shown to be suppressed in hBN-encapsulated monolayers [42]. Here, we focus on the dominant intravalley scattering channels within the K, Λ and K’ valley, cf. Fig. 1(b). First, we illustrate the temperature and density dependence of the EID for the WSe$_2$ monolayer, cf. Fig. 3. As confirmed by earlier experiments [23, 43] and expected from our theory, the EID increases linearly as a function of exciton density $n$, i.e. $\gamma_{\text{KK}} = \gamma_{x-x} n$ with the slope $\gamma_{x-x}$. However, as demonstrated in Fig. 3(b), the slope of the EID is highly temperature-dependent with $\gamma_{x-x} = 3.6 \cdot 10^{-11}$ (1.1 \cdot 10^{-11}) \mu eV cm$^2$ for $T = 300$
FIG. 4. Excitation-induced dephasing of interlayer (IX) and intralayer KK excitons (X) in the MoSe\textsubscript{2}-WSe\textsubscript{2} heterostructure. (a) Density dependence of intralayer and interlayer EID at room temperature. (b) Temperature-dependent EID at the fixed density \(n = 10^{11}\) cm\textsuperscript{-2} revealing \(\gamma_{\text{IX}}(T) \propto T^{-\frac{1}{2}}\).

(50 K). This behavior is governed by the temperature-dependence of the exciton distributions of the involved bright KK and momentum-dark KA and KK’ states (Fig. 3(c)). For low temperatures \((T < 30\text{ K})\), KK’ excitons determine the EID, since most excitons reside in this energetically lowest state, see Table I in the Supplemental Material for the energetic hierarchy of excitonic states in WSe\textsubscript{2}. Above \(T = 50\text{ K}\), the EID is completely dominated by KA excitons reflecting their highest occupation, cf. Fig. 3(c).

Besides the exciton occupation, it is the excitonic Coulomb matrix element that directly determines the efficiency of the underlying exciton-exciton scattering channels, cf. Eq. (4). As illustrated in Fig. 2(a)-(b), the scattering of KK and KA excitons shows the largest matrix element. We ascribe this to the large mass asymmetry of electrons and holes forming KA excitons and enhancing the effective exciton charge. Note that the quantitatively larger values of EID experimentally observed for the WSe\textsubscript{2} monolayer on a sapphire substrate \([23]\) are likely to stem from efficient Auger scattering channels that are suppressed in hBN-encapsulated WSe\textsubscript{2} samples considered in this work \([42]\).

Excitation-induced dephasing in heterostructures. —

Now, we investigate the impact of excitation-induced dephasing on a hBN-encapsulated MoSe\textsubscript{2}-WSe\textsubscript{2} heterostructure. Here, both the EID of the intralayer (X) as well as of the spatially separated interlayer (IX) excitons within the heterostructure is considered. According to a recent DFT study \([44]\), KK excitons are the energetically lowest state in the MoSe\textsubscript{2}-WSe\textsubscript{2} heterostructure, thus the influence of intervalley interlayer KA and KK’ interlayer excitons is expected to be negligible. Qualitatively, the analysis of the density dependence of the intralayer and interlayer EID is similar to the monolayer case, cf. Fig. 4(a). We find a linear density dependence, however with drastically higher slope values of \(\gamma_{\text{IX}}^{\text{hBN}} = 1.2 \cdot 10^{-11}\) meV cm\textsuperscript{-2} and \(\gamma_{\text{IX}}^{\text{SiO}_2} = 2.7 \cdot 10^{-12}\) meV cm\textsuperscript{-2}. Due to the much stronger exciton-exciton interaction for interlayer excitons exhibiting a permanent dipole moment (Fig. 2), the EID is by three orders of magnitude larger compared to the WSe\textsubscript{2} monolayer.

Furthermore, due to the type-II band alignment in the MoSe\textsubscript{2}-WSe\textsubscript{2} heterostructure and the large interlayer energy offset \(\Delta E = 315\) meV between the layers \([20]\), the interlayer exciton state is rendered by far the lowest energetic state. Consequently, exclusively interlayer excitons contribute to the EID at all temperatures. In this particular case, the analytical formula in Eq. (5) can be used to evaluate the EID predicting a \(T^{-\frac{1}{2}}\) temperature-dependence for interlayer excitons, as observed in Fig. 4(b).

Finally, we investigate how the choice of substrate affects the EID. In Fig. 5(a)-(b) the EID is shown as a function of the background dielectric screening \(\epsilon_s\) for two different temperatures. We find a similar behavior for \(T = 50\text{ K}\) and \(T = 300\text{ K}\), namely that the EID in both the monolayer and the heterostructure displays a surprisingly moderate variation due to screening - despite the strong screening dependence of the Coulomb potential itself. This reflects the only moderate decrease of the excitonic Coulomb matrix elements with screening, as illustrated by the inset in Fig. 2(d). The background is that a larger screening also gives rise to an enhanced excitonic Bohr radius, which increases the form factors appearing in the excitonic Coulomb matrix element. Hence, there is a competition between a decreased Coulomb potential \(W_q\) and an increased Bohr radius resulting in a weak overall screening dependence.

By comparing hBN-encapsulated WSe\textsubscript{2} monolayers with WSe\textsubscript{2} on a SiO\textsubscript{2} substrate (marked as dashed vertical lines in Fig. 5), we find a 15% decrease of EID for the
monolayer case at \( T = 50 \) K, but only a 1% decrease at room temperature. This can be understood by considering that the screening has an influence on the excitonic eigenenergies \( \epsilon^b \) and thus changes the exciton occupation in different states \( \mu \). While at room temperature the exciton occupation is spectrally broad and thus the screening-dependent change is relatively small, the situation is different at low temperatures. Here, a shift in excitonic energy when changing the screening from \( \epsilon_s = 2.45 \) to \( \epsilon_s = 4.5 \) has a larger impact on the actual occupation of KA and KK’ states and thus on the efficiency on exciton-exciton scattering and the EID. A similar temperature-dependent screening dependence is also observed for the EID in the heterostructure case.

**Conclusion.**— We have presented a microscopic and fully quantum-mechanic approach on exciton-exciton scattering in 2D materials and related van der Waals heterostructures. In particular, we have investigated the excitation-induced dephasing (EID) taking into account intra- and intervalley as well as intra- and interlayer exciton-exciton scattering channels. We predict an intriguing temperature and screening dependence and explain this by shedding light into the fundamental nature of exciton-exciton scattering. Spatially separated interlayer excitons in heterostructures exhibit a permanent dipole moment and their interaction can be considered as an efficient dipole-dipole coupling resulting in an EID of a few meV. In contrast, intralayer excitons are neutral bosonic quasi-particles and their interaction is only efficient when excitons exhibit a large asymmetry between electron and hole masses. Hence, we find a much more efficient exciton-exciton scattering for momentum-dark intervalley KA excitons in WSe\(_2\) monolayers. The gained microscopic insights can guide future experimental studies on the impact of exciton-exciton scattering on optical properties of excitonic, multi-valley 2D materials.

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Supplementary Material – Excitation-induced dephasing in 2D materials and van der Waals heterostructures

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Note 1. Excitonic Coulomb matrix elements

The starting-point of this work is the exciton-exciton Hamilton operator

\[ H_{x-x} = \frac{1}{2} \sum_{\mu,\nu, Q_1, Q_2, q} V_{q}^{\mu \nu \mu \nu} P_{\mu, Q_1 + q}^{\dagger} P_{\nu, Q_2 - q}^{\dagger} P_{\nu, Q_2} P_{\mu, Q_1}, \]  

(S1)

which is expressed in the excitonic basis [1]. Here, \( P^{(1)} \) are excitonic annihilation (creation) operators, \( Q_1, Q_2 \) are center-of-mass momenta, \( q \) is the momentum transfer, and the excitonic indices \( \mu \) and \( \nu \) can be interpreted as valley indices, since we restrict our calculations to scattering processes (\( \mu \rightarrow \mu, \nu \rightarrow \nu \)) in the lowest energetic 1s state.

In principle, Coulomb exchange interaction processes could also be accounted for, resulting in additional first-order (\( U \propto n^2 \)) and second-order exciton-exciton (\( U \propto n \)) corrections to \( H_{x-x} \) (being the exciton density). However, as explicitly shown for MoS\(_2\) on a SiO\(_2\) substrate [1], both linear and non-linear exchange contributions are small compared to the direct contributions (\( U \sim U \lesssim 0.25V \)).

In this work, we focus on the direct excitonic Coulomb matrix element that reads for TMD monolayers [2]

\[ V_{q}^{\mu \nu \mu \nu}|_{\text{mono}} = V_{q}^{ccc} F_{\mu}(\beta^q) F_{\nu}^{*}(\beta^q) + V_{q}^{vev} F_{\mu}(\alpha^q) F_{\nu}^{*}(\alpha^q) - V_{q}^{ceve} F_{\mu}^{*}(\alpha^q) F_{\nu}^{*}(\beta^q) + F_{\mu}(\alpha^q) F_{\nu}(\beta^q) \]  

(S2)

with the form factors \( F_{\mu}(xq) = \sum_{q_i} \varphi_{\mu,q_{i+1}}(xq) \) determined by the overlap of excitonic wave functions \( \varphi_{\mu,q_{i+1}} \) in valley \( \mu=KK,KK',KK'' \) and the electron/hole mass ratios \( \alpha^\mu = \frac{m_{\epsilon_{\mu}}}{m_{\epsilon_{\mu}^h}}, \beta^\mu = 1 - \alpha^\mu \) with \( m_{\epsilon_{\mu}^h} \) being the corresponding electron (hole) masses at \( \mu=K, \Lambda, K' \). The excitonic wave functions are obtained from solving the Wannier equation using material-specific parameters (masses and dielectric constants) taken from Ref. 3. The intraband Coulomb matrix elements \( V_{q}^{ccc}, V_{q}^{vev} \) and \( V_{q}^{ceve} \) describe direct electron-electron, hole-hole and electron-hole interactions, respectively. These matrix elements can be found in their general form in Ref. 2. For reasons of simplicity, we consider only the long-range part of the Coulomb interaction with a small momentum transfer and thus we can approximate \( V_{q}^{ccc} \approx V_{q}^{vev} \approx V_{q}^{ceve} \approx W_{q} = \frac{V_2}{\varepsilon_q} \) with \( V_2 = \frac{\varepsilon_i^2}{2\varepsilon_0|q|} \) being the two-dimensional bare Coulomb potential with the non-local Keldysh-like [4, 5] monolayer screening \( \varepsilon_q \) given by [6]

\[ \varepsilon_q = \kappa_1 \tanh \left( \frac{1}{2} \alpha_1 d_0 q - \ln \left( \frac{k_1 - k_2}{k_1 + k_2} \right) \right), \]  

(S3)

where \( d_0 \) is the thickness of the TMD layer, and \( \kappa_i = \sqrt{\varepsilon_i^\parallel}, \alpha_i = \sqrt{\varepsilon_i^\parallel/\varepsilon_i^\perp} \) being formed from the perpendicular and parallel components of the dielectric tensor of the considered monolayer \( (i = 1) \) and the environment \( (i = 2) \). Material-specific values on these parameters can be found in Ref. 3.

Within the approximation of the intraband matrix elements introduced above, the monolayer excitonic Coulomb matrix element reduces to

\[ V_{q}^{\mu \nu \mu \nu}|_{\text{mono}} = \frac{V_2}{\varepsilon_q} \left( F_{\mu}(\beta^q) - F_{\mu}^{*}(\alpha^q) \right) \left( F_{\nu}(\beta^q) - F_{\nu}^{*}(\alpha^q) \right) \]  

(S4)

We find that for \( q \rightarrow 0 \), i.e. for long spatial distances in real space, the interaction vanishes. This is reasonable since, excitons are neutral objects and do not interact at large distances. Furthermore, the strength of the interaction is determined by the electron-hole mass asymmetry of the excitons involved in the scattering process indicating that the fermionic substructure of excitons play a vital role in exciton-exciton scattering. We can gain some more intuition for the excitonic Coulomb matrix element by expressing the difference of form factors in (S4) in real space \( F_{\mu}(\beta^q) - \ldots \)
\[ F^\ast_{\mu}(\alpha^\mu q) = \sum_r \varphi_{\mu}^\ast(r)(e^{i\beta^\mu q \cdot r} - e^{-i\alpha^\mu q \cdot r})\varphi_{\mu}(r) \] and considering small momenta \( q \) yielding \( F_{\mu}(\beta^\mu q) - F^\ast_{\mu}(\alpha^\mu q) \approx -q^2 Q_{\mu \nu_B}^2 \). Here, we introduced the (squared) excitonic Bohr radius \( r_{\mu \nu_B}^2 = |\mu| r^2 |\mu| \). Note that \( \langle |\mu| r |\mu| \rangle = 0 \) since the 1s excitonic wave functions are even functions. We also defined the effective excitonic charge density \( Q_{\mu} = \frac{Q_{h(e)} m^h_{\mu} + Q_{e} m^e_{\mu}}{m^h_{\mu} + m^e_{\mu}} \) (S5), with \( Q_{h(e)} = \pm 1 \). It can now be directly seen that the excitonic Coulomb matrix element is enhanced with increasing Bohr radii:

\[ \nu_{q \mu \nu}^{\mu \nu \mu \nu} |_{\text{mono}} \approx W_q r_{\mu \nu_B}^2 r_{\nu_B}^2 g^4 Q_{\mu} Q_{\nu} \quad \text{ (for small } q \text{)}. \] (S6)

As a direct consequence, when increasing the dielectric background screening, the Coulomb potential \( W_q = \frac{V_0}{\varepsilon q} \) becomes reduced, but at the same time the excitonic Bohr radius increases (due to lower binding energies), resulting in an overall moderate screening dependence of the excitonic Coulomb matrix element.

Now, we turn our attention to intralayer (X) and interlayer (IX) excitons in TMD heterostructures. This requires us to modify the dielectric screening in the excitonic Coulomb matrix elements taking into account that electrons and holes can reside in different layers. First, we consider both excitons involved in the scattering process being different layers. Then, we make use of the intralayer and interlayer dielectric screening functions \( \varepsilon_{l_i}^{c_{l_i}} \) and \( \varepsilon_{l_i}^{c_{l_i}} \) respectively, which are given in Ref. 7. This enables us to deduce the corresponding excitonic Coulomb matrix element for IX excitons. Assuming \( l_1 \neq l_2 \) we generalize (S2) to

\[ \nu_{q \mu \nu}^{\mu \nu \mu \nu} |_{\text{IX-X}} = V_{q c_i c_i}^{c_i c_i c_i} F_{\mu}(\alpha^\mu q) F_{\nu}(\alpha^\nu q) + V_{q v_1 v_2 v_2 v_1}^{v_1 v_2 v_2 v_1} F_{\mu}(\beta^\mu q) F_{\nu}(\alpha^\nu q) - V_{q v_1 v_2 v_1 v_1}^{v_1 v_2 v_1 v_1} F_{\mu}(\beta^\mu q) F_{\nu}(\alpha^\nu q) \] (S7)

with \( V_{q c_i c_i}^{c_i c_i c_i} \approx \frac{V_0}{\varepsilon_q^{c_i}} \), \( V_{q v_1 v_2 v_2 v_1}^{v_1 v_2 v_2 v_1} \approx \frac{V_0}{\varepsilon_q^{v_1}} \), \( V_{q v_1 v_2 v_1 v_1}^{v_1 v_2 v_1 v_1} \approx \frac{V_0}{\varepsilon_q^{v_2}} \), where we introduced \( V_q = 2V_0 \) [7]. Note that the products of form factors are weighted differently depending on which layer the electrons/holes reside in. As a consequence, in contrast to the monolayer case the interaction is non-zero even for \( q \to 0 \) and reads

\[ \nu_{q \mu \nu}^{\mu \nu \mu \nu} |_{\text{IX-X}} = \frac{d_0 c_0^2}{\epsilon_0} \left( \frac{1}{\epsilon_{l_1}} + \frac{1}{\epsilon_{l_2}} \right) \quad \text{ (for small } q \text{)} \] (S8)

with \( d_0 \) being the layer thickness (here assumed to be equal for both layers), and \( \epsilon_{l_i}, i = 1, 2 \) denoting the dielectric constants for the individual layers. We can now also consider interlayer-intralayer exciton scattering yielding

\[ \nu_{q \mu \nu}^{\mu \nu \mu \nu} |_{\text{IX-X}} = V_{q c_i c_i}^{c_i c_i c_i} F_{\mu}(\alpha^\mu q) F_{\nu}(\alpha^\nu q) + V_{q v_1 v_2 v_1 v_1}^{v_1 v_2 v_1 v_1} F_{\mu}(\beta^\mu q) F_{\nu}(\alpha^\nu q) - V_{q v_1 v_2 v_2 v_1}^{v_1 v_2 v_2 v_1} F_{\mu}(\beta^\mu q) F_{\nu}(\alpha^\nu q) \] (S9)

In the limiting case of small \( q \) we find

\[ \nu_{q \mu \nu}^{\mu \nu \mu \nu} |_{\text{IX-X}} \approx \frac{d_0 c_0^2}{2\epsilon_0} \left( \frac{1}{\epsilon_{l_1}} + \frac{1}{\epsilon_{l_2}} \right) \quad \text{ (for small } q \). \] (S10)

As performed in detail in the monolayer case, we may also expand the interlayer/intralayer excitonic Coulomb matrix elements in terms of Bohr radii, leading to the same conclusions as above, namely that the matrix elements are enhanced by increased Bohr radii. However, in contrast to the monolayer case, the matrix elements do not scale with \( q \) for small momenta, but exhibit a constant value as a consequence of the permanent dipole moment of interlayer excitons.
Note 2. Excitation-induced dephasing

We start from the simplified exciton-exciton Hamiltonian in (S1) and determine the equation of motion for the excitonic polarisation \( p_{\mu,Q} = \langle P_{\mu}^i P_{\mu Q}^i \rangle \). The evolution of time-dependent quantum-mechanical operators is governed by the Heisenberg equation of motion \( i\hbar \partial_t \langle \cdot \rangle = \{ [\cdot, H], [\cdot, H] \} \) [8] resulting in

\[
- i\hbar \partial_t p_{\mu,Q} = \epsilon Q p_{\mu,Q} + \sum_{\nu Q_1 \neq q} V_{\mu \nu \nu}^{\mu \nu} (P_{\mu,Q+q}^i P_{\nu,Q_1-q}^i P_{\nu Q_1}^i) c^{\dagger}.
\]  

(S11)

Here we only consider the correlated part \( \langle P_{\mu,Q+q}^i P_{\nu,Q_1-q}^i P_{\nu Q_1}^i \rangle_c \) and neglect single-particle factorizations. It is also important to note that we treat the excitonic operators \( P_{\mu}^i \) as bosonic operators, i.e. \( [P_{\mu}, P_{\nu}^i] = \delta_{\mu, \nu} \). In the high excitation regime, the fermionic substructure of excitons has to be taken into account when evaluating the equations of motion and \textit{cobosonic} commutator relations have to be used. However, our Hamiltonian is constructed in such a way that it includes fermionic correction terms compensating for the fact that we use the bosonic commutator relations to derive the equations of motion. For instance, it can be shown that the difference between applying the bosonic commutator and a cobosonic commutator to the direct electron-electron and hole-hole contributions becomes manifest only in different prefactors [2]. The electron-hole scattering part of the excitonic Hamiltonian was verified by explicitly evaluating the equation of motion for the polarisation in the electron-hole picture and transforming the result into the excitonic picture up to the second-order in excitonic operators and comparing with the corresponding result obtained using the excitonic Hamiltonian.

Since we want to go beyond the Hartree-Fock approximation in this work, we derive an equation of motion for the appearing new quantity \( S_{Q+q, Q_1-q}^{\mu \nu} \) yielding

\[
i\hbar \partial_t S_{Q+q, Q_1-q}^{\mu \nu} = \Delta \epsilon S_{Q+q, Q_1-q}^{\mu \nu} + \sum_{\sigma} \left( \nu^{\sigma \mu} Q_{1}^{\mu \nu} - \nu^{\sigma \mu} Q_{1-q}^{\mu \nu} \right) \left( N_{Q_1-q+q}^{\nu} N_{Q+q}^{\mu} - N_{Q_1-q}^{\nu} N_{Q_1}^{\mu} \right) p_{\sigma,Q},
\]

(S12)

with \( \Delta \epsilon = (\epsilon_{Q}^{1}, -\epsilon_{Q}^{1-q} - \epsilon_{Q+q}^{1}) \). Here, we performed correlation expansions of occurring three-operator quantities \( \langle P_{\mu}^i P_{\nu}^i P_{\mu}^i \rangle \) and five-operator quantities \( \langle P_{\mu}^i P_{\nu}^i P_{\mu}^i P_{\nu}^i P_{\mu}^i \rangle \), employing the random-phase approximation (RPA) [8, 9] and neglected higher-order correlations (corresponding to the Born approximation). For instance, the five-operator quantity \( \langle P_{a_1}^i P_{a_2}^i P_{a_3}^i P_{a_4}^i P_{a_5}^i \rangle (a_i, i = 1...5 \text{ being treated as a compound index}) \) can be disentangled by using the following cluster expansion:

\[
\langle P_{a_1}^i P_{a_2}^i P_{a_3}^i P_{a_4}^i P_{a_5}^i \rangle \approx \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle + \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle + \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle + \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle + \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle,
\]

(S13)

with the correlated part \( \langle P_{a_1}^i \rangle \langle P_{a_2}^i \rangle \langle P_{a_3}^i \rangle \langle P_{a_4}^i \rangle \langle P_{a_5}^i \rangle \). Note that this type of factorization procedure only holds if the excitons are treated as pure bosons. Here, we only kept terms proportional to \( \langle P_{a_1}^i \rangle \), which enables us within RPA to find an equation of motion coupling back directly to the excitonic polarisation. Within the RPA, \( \langle P_{\mu}^i P_{\nu}^i \rangle \rightarrow N \) with \( N \) being the exciton occupation. The resulting equation of motion in (S12) can be solved within the Markov approximation [8] yielding

\[
S_{Q+q, Q_1-q}^{\mu \nu} = - i\pi \sum_{\sigma} \left( \nu^{\sigma \mu} Q_{1}^{\mu \nu} + \nu^{\sigma \mu} Q_{1-q}^{\mu \nu} \right) (N_{Q_1-q+q}^{\nu} N_{Q+q}^{\mu} - N_{Q_1-q}^{\nu} N_{Q_1}^{\mu} - N_{Q+q}^{\nu} N_{Q_1}^{\mu} - N_{Q_1-q}^{\nu} N_{Q_1-q+q}^{\mu}) p_{\sigma,Q} \delta(\Delta \epsilon)
\]

(S14)

with \( \Delta \epsilon = \epsilon_{Q}^{1} - \epsilon_{Q}^{1-q} + \epsilon_{Q}^{1-q+q} \). Substituting (S14) into (S11), only taking the diagonal (resonant) part \( \sigma = \mu \), and keeping only the direct scattering term and the linear term in exciton occupation enables us to arrive at a simple equation of motion

\[
\partial_t p_{\mu,Q} |_{H_{x}} = - \gamma_{Q}^{\mu} p_{\mu,Q},
\]

(S15)

where the excitation-induced dephasing is apparent reading

\[
\gamma_{Q}^{\mu} = \frac{\pi}{\hbar} \sum_{\nu Q_1, q} |V_{q}^{\mu \nu}|^2 N_{Q_1}^{\nu} \delta(\epsilon_{Q_1+q}^{\nu} - \epsilon_{Q}^{1} + \epsilon_{Q}^{1-q} - \epsilon_{Q+q}^{1})
\]

(S16)

Here we used the relations \( V_{q}^{\mu \nu} = V_{-q}^{\nu \mu} = (V_{q}^{\mu \nu})^* \) and shifted indices \( Q_1 \rightarrow Q_1 + q \).
Note 3. Energies, masses and Bohr radii

In Table I, we provide the excitonic eigenenergies and masses which are required to evaluate the excitation-induced dephasing in hBN-encapsulated WSe\(_2\) monolayers and the MoSe\(_2\)-WSe\(_2\) heterostructure. Due to the large interlayer energy offset \(\Delta E = 315\) meV in MoSe\(_2\)-WSe\(_2\), KK is rendered by far the lowest energetic state [7]. We also include the corresponding Bohr radii, which enter directly the excitonic Coulomb matrix element expanded around small momenta, cf. Eq. (S6).

| Valley \(\mu = (\mu_h, \mu_e)\) | \(E^\mu\) (meV) | Bohr radius \(r_{\mu,B}\) (nm) | Hole mass \(m_{\mu}^{\text{ch}}\) \((m_0)\) | Electron mass \(m_{\mu}^{\text{el}}\) \((m_0)\) |
|-------------------------------|----------------|------------------------------|-----------------|-----------------|
| KK                           | -169           | 1.72                         | 0.36            | 0.29            |
| KΛ                           | -213           | 1.31                         | 0.36            | 0.6             |
| KK‘                          | -224           | 1.50                         | 0.36            | 0.4             |
| KK (interl. MoSe2-WSe2)      | -407           | 1.59                         | 0.6             | 0.29            |

TABLE I. Effective hole mass \(m_{\mu}^{\text{ch}}\), electron masses \(m_{\mu}^{\text{el}}\), Bohr radius \(r_{\mu,B}\) and excitonic energy \(E^\mu\) in hBN-encapsulated monolayer WSe\(_2\) for different intra- and inter-valley exciton species. The last line also includes data for KK interlayer excitons in the MoSe\(_2\)-WSe\(_2\) heterostructure. Note that \(E^{KK-\text{interl}}\) includes the interlayer energy separation \(\Delta E = 315\) meV. The excitonic energies are obtained from solving the Wannier equation. The effective masses are expressed as fractions of the free electron mass, \(m_0\), and are taken from Ref. 3. The Bohr radius is computed as the average distance between the electron and the hole, i.e. \(r_{\mu,B} = \sqrt{\langle |r|^2 |\mu\rangle}\), with the expectation value taken with respect to the 1s exciton wave functions in the corresponding valley \(\mu = KK, KΛ, KK’\).

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