Quantum Interference Control of Localized Carrier Distributions in the Brillouin Zone

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Using transition metal dichalcogenides as an example, we show that the quantum interference arising in 2- and 3-photon absorption processes can lead to controllable, highly localized carrier distributions in the Brillouin zone. We contrast this with the previously studied 1- and 2-photon absorption, and find qualitatively new features, including changes in the relevance of interband and intraband processes according to the excitation energy. Furthermore, the distribution of excitations arising under certain circumstances in 2- and 3-photon absorption can facilitate the study of far-from-equilibrium states that are initially well-localized in crystal momentum space.

I. INTRODUCTION

Although nearly every technological device is based on systems in far-from-equilibrium states, our understanding of the properties of materials in such a regime is limited [1, 2]. This is the case even for intensively investigated materials such as semiconductors, which are the basis of digital technology. One of the main impediments in the study of materials far-from-equilibrium is the difficulty in creating quantum excitations in a controlled way. For example, we lack good methods for creating one of the simplest types of electronic excitation in a gapped material: the excitation of an electron from a valence to a conduction band at a given crystal momentum. A simple way to excite electrons from one band to another is via optical fields, but they usually excite carriers in almost every location of the Brillouin zone where the photon energy matches the energy difference of the electronic bands. However, because of quantum interference effects, optical fields of different frequencies can be used in combination to excite carriers with more local distributions in the Brillouin zone.

Quantum interference between distinct processes that result in the same transition can lead to localized electronic excitations in crystal momentum space, as different processes interfere constructively in some parts of the Brillouin zone and destructively in others. This interference can be manipulated by varying physically tunable parameters of the system such as a relative phase parameter of the optical fields, and their intensities. Polar distributions of injected carriers in the Brillouin zone lead to charge currents, and such “injected currents” due to the interference of 1- and 2-photon absorption processes (“1+2” injection) have been observed in bulk [3–7] and 2D materials [8–11]. The analogous injection of spin currents has been detected in bulk semiconductors [12–16], and proposed in topological insulators [17]; the injection of spin and valley currents in transition metal dichalcogenides has also been proposed [18].

This quantum interference control (QuIC) of carriers has been exploited to determine the carrier-envelope phase of pulses short enough to contain both the fundamental frequency and its second harmonic [19, 20]; semiconductors with a relatively large band gap are of interest here to allow room temperature operation. Recently the use of interference between 2- and 3-photon absorption processes to inject currents in semiconductors has also been studied both experimentally [21] and theoretically [22]. This “2+3” injection is of special interest for determining the carrier-envelope phase of short pulses, since it can be used even if the frequency spread of the short pulse does not span an octave. Coherent optical frequency combs can be used for studying even more general “n + m” QuIC of carriers in gapped materials.

The detection of these interference processes has typically been by observing the net current they generate, either directly by electrodes [4] or indirectly through the THz radiation resulting from the excitation and subsequent decay of that current [8]. These detection schemes are sensitive only to the first moment of the carrier distribution in the Brillouin zone, and that is all that has been typically calculated. However, recent advances in time-resolved ARPES [23] offer the promise of detecting carrier distributions in the Brillouin zone as a function of time. This would yield unparalleled insight into the relaxation processes of such excitation distributions injected by QuIC, where carriers can be placed in regions of the Brillouin zone far from those occupied by equilibrium or near-equilibrium carrier distributions. It is also particularly convenient to use QuIC in combination with such pump-probe spectroscopic techniques for if one were to use a pumping field that facilitates QuIC, one can set the system in a far-from-equilibrium state having a distribution of electronic excitations that is well-localized in crystal momentum space, and subsequently study the decay of this distribution via the probe...
pulsed. So detailed theoretical studies of the injected carrier distributions, and ultimately of their subsequent dynamics, are now in order.

In this first communication along these lines, we study the initial carrier distributions in the transition metal dichalcogenide (TMD) WSe$_2$ due to 1+2 and 2+3 injection. An important result is that 2+3 injection can lead to more localized carrier distributions in the Brillouin zone than 1+2 injection, moving further towards the goal of coherent control strategies that act as effective “tweezers in the Brillouin zone” for the placement of carriers where desired. The outline of the paper is as follows: We begin in Section II by introducing a generic single-particle Hamiltonian, where the vector potential is included via minimal coupling, and derive expressions for the first-, second-, and third-order perturbative coefficients. Using these coefficients we illustrate the origin of QuIC. In Section III we introduce the quantities of interest, namely the electron excitation (carrier injection) rate, and the current injection rate. Following this, in Section IV, we introduce the model Hamiltonian for TMDs and use this system as a platform to compare features of different orders of photon absorption processes. In the sections following this we analyse our findings; Section V contains the distributions of electronic excitations for various polarizations of incident light, and in Section VI we plot the dependence of the carrier and current response tensors on the excitation energy.

II. OPTICAL INJECTION RATES

We investigate the optical excitation of electrons by way of time-dependent perturbation theory (TDPT), using a fully quantized Hamiltonian $\mathcal{H}(t)$ that follows from a single particle Hamiltonian density of the form

$$
\mathcal{H}(x, p; t) = \frac{1}{2m} \left( p - \frac{e}{c} A(t) \right)^2 + \mathcal{H}_S(x, p - \frac{e}{c} A(t)) + \mathcal{H}_{lo}(x),
$$

(1)

where $e = -|e|$ is the electronic charge, $x$ and $p$ are position and momentum operators, $\mathcal{H}_S$ is the spin-orbit term, and $\mathcal{H}_{lo}(x)$ is the periodic lattice potential energy. We have chosen a gauge in which the time-dependent scalar potential is zero, and have assumed the external electromagnetic field can be approximated as uniform, with an electric field $E(t)$ described solely by the vector potential $A(t)$. We only consider electrons and holes injected at high enough energies to lead to currents, and so bound exciton states are not relevant. The electron-hole interaction in the ionized excitons that result can lead to phase shifts in the injected currents at excitation energies close to the band gap [24]; we ignore those here, as well as other effects of electron-electron interactions. The $O(A(t)^2)$ term arising in (1) is solely a function of time and adds a global phase to the energy eigenstates, which has no consequence on the expectation values we compute. Thus the interaction term takes the form $V_{ext}(t) = -e \mathbf{v} \cdot \mathbf{A}(t)$, where $\mathbf{v} = \frac{1}{2} [\mathcal{H}_S(x, p)]$ is the velocity operator; this holds for any unperturbed single-particle Hamiltonian that is at most quadratic in the momentum. The only experimentally accessible parameters within $\mathcal{H}(t)$ enter through this external interaction; namely the intensity, polarization, and phase of the optical fields. It is therefore these parameters that can then be varied to tune the quantum interference between excitation processes.

In second quantized notation the full Hamiltonian can be written as $\mathcal{H}(t) = \mathcal{H}_0 + V_{ext}(t)$, where

$$
\mathcal{H}_0 = \sum_{nk} \hbar \omega_n(k) a_{nk}^\dagger a_{nk},
$$

$$
V_{ext}(t) = \sum_{nmk} \gamma_{nm}(k, t) a_{nk}^\dagger(t) a_{mk}(t).
$$

(2)

The crystal momentum wave vectors, $k$, are summed over the first Brillouin zone, $n$ and $m$ label bands, and $\gamma_{nm}(k, t) = -e \mathbf{v}_{nm}(k) \cdot \mathbf{A}(t)$, with $\mathbf{v}_{nm}(k) = (\langle n | k | m \rangle \omega_{nm})$. We express (2) in the basis of eigenstates of $H_0$, where $a_{nk}^\dagger | vac \rangle = | mk \rangle$ indicates a Bloch state of band $m$ with crystal momentum $hk$ and energy $\hbar \omega_m(k)$. We take a vector potential of the form

$$
\mathbf{A}(t) = \sum_\alpha a_{\alpha} e^{-i(\omega_{\alpha} + \epsilon t)\gamma} = -\sum_\alpha \frac{ic}{\omega_\alpha} E_{\alpha} e^{-i(\omega_{\alpha} + \epsilon t)\gamma},
$$

(3)

where $\epsilon \rightarrow 0^+$ describes the adiabatic turning on of the optical fields from $t = -\infty$, and for $n + m$ injection we sum over frequencies $\omega_\alpha = \pm \Omega/n, \pm \Omega/m$, where $\hbar \Omega$ identifies the total transition energy. To keep track of the relative phases and the polarizations of the different frequency components we write $E_{\omega_\alpha} = E_{\alpha} e^{i\phi_{\omega_\alpha}}$ $\hat{e}_{\omega_\alpha}$, where $E_{\alpha}$ and $\phi_{\omega_\alpha}$ are real valued and $\hat{e}_{\omega_\alpha}$ is a polarization vector satisfying $\hat{e}_{\omega_\alpha} \cdot \hat{e}_{\omega_{\alpha'}} = 1$.

The implementation of perturbation theory for problems of this type has been previously discussed [25, 26]; here we simply summarize the results. Under the time-evolution operator $U(t)$ associated with $\mathcal{H}(t)$, the full ket $| \psi(t) \rangle$ is given by

$$
| \psi(t) \rangle = U(t) | gs \rangle = \gamma_0(t) | gs \rangle + \sum_{cvk} \gamma_{cv}(k, t) | cvk \rangle + ...
$$

(4)

where $| gs \rangle$ denotes the ground state of the unperturbed Hamiltonian, where all conduction bands (labeled $c$) are empty and all valence bands (labeled $v$) are occupied. The excited states of interest are $| cvk \rangle \equiv a_{ck}^\dagger a_{vk} | gs \rangle$, and the coefficients

$$
\gamma_{cv}(k, t) = \langle cvk | U(t) | gs \rangle
$$

(5)

must be computed. The perturbative calculation gives

$$
\gamma_{cv}(k, t) = \mathcal{R}_{cv}(k) e^{-i(\Omega + \epsilon t)\gamma} \frac{\rho_{cv}(k)}{\Omega - \omega_{cv}(k) + i\epsilon},
$$

(6)
where \( \omega_c(k) = \omega_c(k) - \omega_v(k) \) is the frequency difference between the bands \( c \) and \( v \) at \( k \), and where \( R_{cv}(k) \) is the sum of terms \( R_{cv}^{(N)}(k) \); these terms are transition amplitudes arising at different order, \( N \), of TDPT. Here we are interested in the amplitudes \( R_{cv}^{(1)}(k) \), \( R_{cv}^{(2)}(k) \), and \( R_{cv}^{(3)}(k) \), which we identify with 1-, 2-, and 3-photon absorption, respectively. We find

\[
R_{cv}^{(N)}(k) = \sum_{\alpha,...,\beta} R_{cv}^{(N)\alpha\beta}(k; \omega_\alpha, ..., \omega_\beta) E^a_{\omega_\alpha} \cdots E^b_{\omega_\beta},
\]

where the sum is over both frequencies and Cartesian components, the latter indicated by superscript indices; at \( N^{th} \) order perturbation theory, there are \( N \) frequencies in the list \( (\omega_2, ..., \omega_N) \) that add to \( \Omega \), and \( N \) Cartesian components in the list \( a \cdots b \). The amplitudes \( R_{cv}^{(N)}(k) \) of interest in are indicated in Fig. 1 for the pairs of absorption processes occurring together that we will consider. For the lowest few \( N \) we find

\[
R_{cv}^{(1)}(k; \omega_\alpha) = \frac{i e}{\hbar \omega_\alpha} \psi_{\alpha v}(k),
\]

\[
R_{cv}^{(2)ab}(k; \omega_\alpha, \omega_\beta) = -\frac{e^2}{\hbar^2 \omega_\alpha \omega_\beta} \sum_c \frac{\psi_{\alpha c}^{(2)}(k) \psi_{cv}^{(2)}(k)}{\omega_\alpha - \omega_{cv}(k)} - \sum_c \frac{\psi_{\beta c}^{(2)}(k) \psi_{cv}^{(2)}(k)}{\omega_\beta - \omega_{cv}(k)} + \sum_c \frac{\psi_{\alpha c}^{(2)}(k) \psi_{\beta c}^{(2)}(k)}{\omega_\alpha - \omega_{cv}(k)} + \sum_c \frac{\psi_{\beta c}^{(2)}(k) \psi_{\alpha c}^{(2)}(k)}{\omega_\beta - \omega_{cv}(k)},
\]

\[
R_{cv}^{(3)ab}(k; \omega_\alpha, \omega_\beta, \omega_\delta) = \frac{i e^3}{\hbar^3 \omega_\alpha \omega_\beta \omega_\delta} \left[ \sum_{c,c'} \frac{\psi_{\alpha c}^{(3)}(k) \psi_{cv}^{(3)}(k)}{\omega_\alpha - \omega_{cv}(k)} - \sum_{c,c'} \frac{\psi_{\beta c}^{(3)}(k) \psi_{cv}^{(3)}(k)}{\omega_\beta - \omega_{cv}(k)} + \sum_{c,c'} \frac{\psi_{\delta c}^{(3)}(k) \psi_{cv}^{(3)}(k)}{\omega_\delta - \omega_{cv}(k)} \right].
\]

**III. CARRIERS AND CURRENTS**

We are interested in the injection rates of both conduction electrons and current density arising from the electronic placement in the Brillouin zone just after the external fields have been removed. The operators corresponding to the density of electrons in the conduction bands and the total current density are given respectively by

\[
n_e(t) = \frac{1}{L^D} \sum_{c,k} a_{c,k}^\dagger a_{c,k},
\]

\[
J(t) = \frac{1}{L^D} \sum_{m,n,k} \epsilon_{mn}(k) a_{n,k}^\dagger(t) a_{mk}(t),
\]

where \( L \) is a normalization length, \( D \) is the spatial dimension of the system, and in the latter term \( n \) and \( m \) range over all bands. To investigate the distribution of electronic excitations in crystal momentum space arising from various photon absorption processes, we resolve the injected densities through the Brillouin zone; for a general operator \( M(t) \), which here could be \( n_e(t) \) or \( J(t) \), we find we always have

\[
\langle \psi(t) | M(t) | \psi(t) \rangle = \int \frac{d^D k}{(2\pi)^D} \langle M(k; t) \rangle,
\]

where \( M(t; k) \) is the “density” in the Brillouin zone associated with the operator \( M(t) \). It will be these densities that we later plot, primarily \( \frac{1}{L^D} \langle n_e(t) \rangle_{n,m} \), and are the main focus of this paper. It also happens that under our approximations [26] \( \langle M(k; t) \rangle \) is independent of time.
A. 1+2 photon absorption

Consider a system with a direct band gap $E_g$ as indicated in Fig. 1a, with an incident optical field composed of frequencies $\omega$ and $2\omega$, where $\hbar \omega < E_g < 2\hbar \omega$. In such a system we require a total energy of at least $E_g$ to excite electrons from valence to conduction bands, which can be satisfied both by absorption of a single photon of energy $2\hbar \omega$ and by the absorption of two photons each of energy $\hbar \omega$. We illustrate below how the interference of these two excitation pathways gives rise to an injected current density in the system. Contributions to the total conduction electron density injection rate have the form

$$\frac{d}{dt} \langle n_i \rangle = \xi^{ab}_1 (2\omega) E_{-2\omega} E_{2\omega},$$

$$\frac{d}{dt} \langle n_i \rangle^{(1+2)}(2\omega) = \xi^{(1+2)}_2 (2\omega) E_{-2\omega} E_{2\omega} + c.c.,$$

where, in the above and below, we use the subscript $i$ in $\frac{d}{dt} \langle n_i \rangle^{(1+2)}(n,m)$ to denote a contribution to the total rate that arises solely from the interference of excitation processes.

For a general response coefficient, which might for example be $\xi^{ab}_1 (2\omega)$, or $\xi^{(1+2)}_2 (2\omega)$, or $\xi^{abde}_2 (2\omega)$, we always find there to be an associated Brillouin zone “density” (analogous to the densities associated with operators) that, when weighted by factors of the fields and summed appropriately, give those operator densities as identified in (13). For example, we have

$$\frac{d}{dt} \langle n_i \rangle^{(1+2)}(2\omega;k) = 2\pi \sum_{c,v} R^{(1+2)}_{cv}(k; 2\omega)^* \times R^{(1+2)}_{cv}(k; 2\omega) \delta (2 \omega - \omega_c(k)).$$

Adopting this notation, we identify

$$\xi^{ab}_1 (2\omega;k) = 2\pi \sum_{c,v} R^{(1)}_{cv}(k; 2\omega)^* \times R^{(1)}_{cv}(k; 2\omega) \delta (2 \omega - \omega_c(k)).$$

$$\xi^{abde}_2 (2\omega;k) = 2\pi \sum_{c,v} R^{(2)}_{cv}(k; \omega, \omega)^* \times R^{(2)}_{cv}(k; \omega, \omega) \delta (2 \omega - \omega_c(k)).$$

while the current density injection rate is given by

$$\frac{d}{dt} \langle \mathcal{J}^{a}_+ \rangle^{(1+2)} = \eta^{abde}_1 (2\omega) E_{-2\omega} E_{2\omega} + c.c.,$$

with

$$\eta^{abde}_1 (2\omega;k) = 2\pi \sum_{c,v} e^{[v^a_{cv}(k) - v^a_{cv}(k)]} R^{(1+2)}_{cv}(k; \omega, \omega)^* \times R^{(2)}_{cv}(k; \omega, \omega) \delta (2 \omega - \omega_c(k)).$$

for systems having current injected only because of the interference of excitation processes; namely those systems for which $\eta^{abde}_1 = \eta^{abdef}_2 \equiv 0$.

B. 2+3 photon absorption

Next suppose the optical field is composed of frequencies $\omega$ and $3\omega/2$, with $2\hbar \omega < E_g < 3\hbar \omega$. Then 2- and 3-photon absorption processes can promote electrons from valence to conduction bands, and also interfere; see Fig. 1b. The contributions to the conduction electron density injection rate are

$$\frac{d}{dt} \langle n_i \rangle^{(2+3)}(3\omega) = 2\pi \sum_{c,v} R^{(2)}_{cv}(k; 3\omega, \omega)^* \times R^{(2)}_{cv}(k; 3\omega, \omega) \delta (3 \omega - \omega_c(k)).$$

$$\xi^{abdef}_2 (3\omega;k) = 2\pi \sum_{c,v} R^{(3)}_{cv}(k; \omega, \omega, \omega)^*$$

where

$$\xi^{abedef}_3 (3\omega;k) = 2\pi \sum_{c,v} R^{(3)}_{cv}(k; \omega, \omega, \omega)^*$$

and

$$\xi^{abdef}_3 (3\omega;k) = 2\pi \sum_{c,v} R^{(3)}_{cv}(k; \omega, \omega, \omega)^*$$

with

$$\eta^{abdef}_2 (3\omega;k) = 2\pi \sum_{c,v} e^{[v^a_{cv}(k) - v^a_{cv}(k)]} R^{(2)}_{cv}(k; \omega, \omega, \omega)^* \times R^{(2)}_{cv}(k; 3\omega, \omega, \omega) \delta (3 \omega - \omega_c(k)).$$

Again, we consider systems satisfying $\eta^{abdef}_2 \equiv 0$. The final expression is

$$\frac{d}{dt} \langle \mathcal{J}^{a}_+ \rangle^{(2+3)} = \eta^{abdef}_2 \equiv 0.$$
C. 1+3 photon absorption

Finally, consider an incident optical field consisting of frequencies \( \omega \) and \( 3\omega \), with \( 2\hbar\omega < E_g < 3\hbar\omega \); see Fig. 1c. The contributions to the conduction electron density injection rate have the form

\[
\frac{d}{dt} \langle n_c \rangle_1 = \xi_1^{ab}(3\omega) E_{-\omega}^a E_{3\omega}^b + c.c.,
\]

\[
\frac{d}{dt} \langle n_c \rangle_{1+3} = \xi_1^{abde}(3\omega) E_{-\omega}^a E_{-\omega}^d E_{3\omega}^e E_{3\omega}^f + c.c.,
\]

where the densities of the response coefficients in the Brillouin zone are given by

\[
\xi_1^{ab}(3\omega; k) = 2\pi \sum_{cv} R_{cv}^{(1)ab}(k; 3\omega) \delta(3\omega - \omega_v(k)),
\]

\[
\xi_1^{abde}(3\omega; k) = 2\pi \sum_{cv} R_{cv}^{(3)abde}(k; \omega, \omega, \omega) \delta(3\omega - \omega_v(k)),
\]

\[
\xi_3^{abdef}(3\omega; k) = 2\pi \sum_{cv} R_{cv}^{(3)abdef}(k; \omega, \omega, \omega) \delta(3\omega - \omega_v(k)).
\]

Similarly, the current density injection rate is given by

\[
\frac{d}{dt} \langle \mathcal{J} \rangle_{1+3} = \eta_1^{abdef}(3\omega) E_{-\omega}^a E_{-\omega}^d E_{3\omega}^e E_{3\omega}^f + c.c.,
\]

where

\[
\eta_1^{abdef}(3\omega; k) = 2\pi \sum_{cv} \left| v_{cv}^{a}(k) - v_{cv}^{b}(k) \right| R_{cv}^{(1)abdef}(k; \omega, \omega, \omega) \delta(3\omega - \omega_v(k)).
\]

and again we are restricting ourselves to systems satisfying the condition \( \eta_1^{ab} = \eta_3^{abdefg} \equiv 0 \).

D. Qualitative functional behaviour of the response coefficients

Before evaluating these expressions for a particular model, we note some general features that can be expected from any system with a direct band gap in which electron-electron and electron-phonon interactions are neglected, as well as possible small contributions from other bands.

In the 1-photon absorption amplitude (8) there is a single interband velocity matrix element, \( v_{cv}(k) \), which is in general non-zero for all \( k \) values. In the 2-photon absorption amplitude (9) there are products of inter- and intraband velocity matrix elements, while the 3-photon absorption amplitude contains terms involving one interband and two intraband elements, as well as terms involving three interband elements.

The structure of these terms is vital to understanding the variation of the absorption amplitudes through the Brillouin zone. Unlike the interband velocity matrix element, the intraband matrix elements \( v_{cv}(k) \) are zero at the conduction (valence) band minima (maxima), since they are directly related to the slope of the bands at that \( k \)-point; in particular, they both vanish at the band gap, and have a much more significant crystal momentum dependence than the interband matrix elements. Thus the 2-photon absorption amplitudes have more structure in the Brillouin zone than do the 1-photon amplitudes.

This is even more dramatic for the component of the 3-photon amplitude involving two intraband terms, which also vanishes at a band extremum, but varies more quickly in \( k \) than do the 2-photon amplitudes due to the appearance of two diagonal matrix elements \( v_{cv}(k) \). In contrast to the 2-photon amplitude, the 3-photon amplitude has a component composed entirely of interband matrix elements that is non-zero at the band gap, and generally has a slow variation in the Brillouin zone, reminiscent of the 1-photon absorption amplitude.

In this way, the 3-photon absorption process contains qualitative features of both the 1- and 2-photon processes.

Since the 2- and 3-photon absorption amplitudes can generally be expected to exhibit much more structure in the Brillouin zone than the 1-photon amplitude, it is not surprising that carriers injected through a 2+3 absorption process, where there is interference between the 2- and 3-photon absorption amplitudes, can be more localized in the Brillouin zone than those injected from a 1+2 absorption process, involving interference between 1- and 2-photon absorption amplitudes.

Finally, for a material with centre-of-inversion symmetry all full response coefficients described by odd rank tensors vanish. For such a material \( \xi_{1+2}^{(1+2)\omega} \) and \( \xi_{1+3}^{(1+3)\omega} \) are identically zero. That is, in both 1+2 and 2+3 photon absorption there is no interference between the contributing processes that can lead to QuIC of the total number of electron-hole pairs created. Nonetheless, \( \xi_{1+2}^{(2+2)\omega} \) and \( \xi_{1+3}^{(2+3)\omega} \) are not identically zero. That is, at a particular point in the Brillouin zone there can be interference between the contributing processes, with (say) more electron-hole pairs created at a particular \( k_{\omega} \) than at \(-k_{\omega}\), and indeed it is this interference that leads to the injected current described by the even rank tensor \( \eta_{1+2}^{(2+2)\omega} \) (for 1+2 PA) or \( \eta_{1+3}^{(2+3)\omega} \) (for 2+3 PA).

The situation for 1+3 photon absorption is qualitatively different. For a material with centre-of-inversion symmetry there is no QuIC leading to an injected current, as the tensor \( \eta_{1+3}^{(1+3)\omega} \) that describes it is of odd rank. However, the tensor \( \xi_{1+3}^{(1+3)\omega} \) that describes QuIC related to the total number of electron-hole pairs injected is of even rank, and so this QuIC
survives.

We now illustrate these features with a calculation using a model for TMDs.

IV. LOW-ENERGY MODEL FOR TRANSITION METAL DICHLORIDE

Optical fields are only able to excite electrons near the Fermi surface, and so it is solely the low-energy physics of the system that is relevant to study such a perturbation. We therefore consider an effective theory defined in the regions near the valleys, $K$ and $K'$, that describes the low-energy excitations of the system; we let $k$ indicate the displacement in the Brillouin zone from the nearby valley, writing

$$k = k_x \hat{x} + k_y \hat{y} = k (\cos \theta + \sin \theta \sin \theta),$$  

where $k \equiv |k|$ and $\theta$ is the angle that $k$ makes from the $\hat{x}$ axis. We adopt an exactly solvable model Hamiltonian used frequently in the literature [27–30], that includes terms allowed by the symmetry of the lattice, and explicitly retains the intratomic spin-orbit coupling term:

$$\mathcal{H}_0(k) = \hbar \overline{A} \left( k_x \sigma_z \otimes \sigma_z \otimes S_0 + k_y \tau_0 \otimes \sigma_y \otimes S_0 \right)$$

$$+ \frac{\hbar \Delta}{2} \left( \tau_0 \otimes \sigma_z \otimes S_0 \right) + \hbar k (\tau_0 \otimes (\sigma_0 - \sigma_z) \otimes \sigma_z),$$

where the components of $\tau, \sigma$, and $s$ are the usual Pauli matrices, and with the index 0 referring to the $2 \times 2$ identity matrix over the appropriate Hilbert space. For $\tau$ that Hilbert space corresponds to the valley degree of freedom associated with the massive Dirac points, for $\sigma$ with the pseudospin degree of freedom associated with the inequivalent sublattice sites, and for $s$ with the spin degree of freedom.

In general $\mathcal{H}_0(k)$ is represented by an $8 \times 8$ matrix; but the valleys are not coupled, and about each valley the spin degrees of freedom also decouple. So a valley-dependent Hamiltonian can be written involving two $2 \times 2$ matrices. We thus solve a general $2 \times 2$ eigenvalue equation for an input $\{\tau, s\}$, which we use to specify the valley and spin for which we are solving: $\tau = 1(-1)$ corresponds to the $K$ ($K'$) valley, and $s_0 = 1(-1)$ to spin component in the $z$ direction being up (down):

$$\mathcal{H}_\tau(k) = \hbar \sigma_z \sigma_0 + \hbar d_{\tau \sigma}(k) \cdot \sigma,$$  

where $\sigma = \lambda \tau \tau_x, d_{\tau \sigma}(k) = \Xi k_x \hat{x} + \Xi k_y \hat{y} + \Delta_{\tau \sigma} \hat{z},$ and $\Delta_{\tau \sigma} = (\Delta - \lambda \tau \tau_x)/2$. Here $\Xi$ is related to the hopping integral from site to site, $\hbar \Delta$ is the band gap energy if SOI were to be neglected, and $\lambda$ characterizes the spin-orbit coupling; their values are given in Table I for WSe$_2$. Note that $\Delta_{\tau \sigma} > 0$.

One can diagonalize the Hamiltonian (29) to find energies $\varepsilon^c_{\sigma}(k) = \hbar \sigma \tau \sigma_0 (\sigma_{\tau \sigma}(k))$ (see Fig. 2a and 2b) for a given $\{\tau, s\}$, leading to a frequency difference

$$\omega^{c}_{\tau \sigma}(k) = 2|\sigma_{\tau \sigma}(k)|$$  

between the bands $c$ and $v$ associated with $\{\tau, s\}$ at $k$; note that

$$|\sigma_{\tau \sigma}(k)|^2 = \Xi^2 k^2 + \Delta_{\tau \sigma}^2$$

is independent of $\theta$, even though $\sigma_{\tau \sigma}(k)$ is not, and the frequency difference function for a particular spin in one valley is equal to the frequency difference function for the opposite spin in the other valley. This is due to the combination of time-reversal symmetry and because $\varepsilon^{c}_{\tau \sigma}(k) = -\varepsilon^{v\tau}(k)$, which together imply $\varepsilon^{c}_{\tau \sigma}(k) = -\varepsilon^{v\tau}(k)$.

| $\hbar \Xi$ | $\hbar \lambda$ | $\hbar \Delta$ | $E_{0a}(\nu)$ | $E_{0b}(\nu)$ |
|---|---|---|---|---|
| 3.9 eV | 0.46 eV | 1.6 eV | $2 \times 10^4$ | $1.3 \times 10^6$ | $2 \times 10^7$ |

TABLE I: Model parameters for WSe$_2$ and upper bound of optical field amplitudes for single colour absorption.

The only model dependent parameters that arise in the perturbative expansion of the transition coefficient $\gamma_{cv}(k, \Omega)$ are related to the velocity matrix elements [18]. Writing $\hat{e}_z = (\hat{x} + i \hat{y})/ \sqrt{2} = \hat{e}^c_z$, the interband velocity matrix elements are given by

$$v_{cc}(k) = \frac{\Xi e^{-i k \tau \sigma}}{\sqrt{2}} \left[ e^{-i \theta} \left( \frac{\tau \Delta_{\tau \sigma}}{|d_{\tau \sigma}(k)|} - 1 \right) \hat{e}^c_z + e^{i \theta} \left( \frac{\tau \Delta_{\tau \sigma}}{|d_{\tau \sigma}(k)|} + 1 \right) \hat{e}^c_z \right]$$

$$= -i \Xi e^{-i k \tau \sigma} \left[ -\sin \theta + i \left( \frac{\tau \Delta_{\tau \sigma}}{|d_{\tau \sigma}(k)|} \right) \cos \theta \right] \hat{x}$$

$$+ \left( \cos \theta + i \left( \frac{\tau \Delta_{\tau \sigma}}{|d_{\tau \sigma}(k)|} \right) \sin \theta \right) \hat{y}$$

while the intraband velocity matrix elements are given by

$$v_{cc}(k) = -v_{vv}(k) = \frac{\Xi^2 k}{|d_{\tau \sigma}(k)|}.$$
which follow immediately from (32,33). We also introduce $k_{rs}(m\omega)$, the crystal momentum at which $\delta(m\omega - \omega_{cv}^Z(k))$ is satisfied, and whose magnitude is given by

$$k_{rs}(m\omega) = \Xi^{-1} \sqrt{\left(\frac{m}{2}\omega\right)^2 - \Delta_z^2}, \quad (36)$$

The velocity matrix elements are plotted in Fig. 2c along the direction $\theta = 0$ in the Brillouin zone. In the variation of these matrix elements through the Brillouin zone, and in the way they combine in the amplitudes (34), one can easily identify the different qualitative nature of the absorption amplitudes, as discussed in Section III-D. While the TMDs lack centre-of-inversion symmetry, for the configuration of optical fields at normal incidence to which we restrict ourselves there are electric fields only in the $\hat{x}$ and $\hat{y}$ directions, and we consider only current injections in the plane defined by those two vectors. For this restricted set of “in plane” Cartesian components the TMD response coefficients do exhibit the selection rules that would follow from centre-of-inversion symmetry: All third rank tensors vanish, QuIC of injected current is possible only for $1 + 2$ and $2 + 3$ absorption, and QuIC of the number of injected electron-hole pairs is possible only for $1 + 3$ absorption. Expressions for the full set of non-vanishing response coefficients (14,17,19,21,23) for this model are given in Appendix A.

V. ELECTRONIC DISTRIBUTION IN THE BRILLOUIN ZONE

In what follows we focus primarily on an excitation energy of $\hbar\Omega = 1.5$ eV, which should be assumed unless otherwise specified. To display the location of injected carriers in the Brillouin zone, we plot $\frac{d}{dt} \langle n_{cv}(k) \rangle_{n+m}$ for $n + m$ being $1 + 2$, $2 + 3$, and $1 + 3$. These quantities are found by weighting the previously found densities of the carrier injection rate coefficients, $\xi(\Omega; k)$, by factors of the fields, and summing them appropriately; this was described in detail in Section III.

Since our calculations are done at the perturbative level, the field amplitudes and pulse lengths must be such that only a small fraction of carriers in any region of the valence band are excited into the conduction band. Taking a nominal figure of 5% for this limit, the rough upper bounds for optical field amplitudes corresponding to 1-, 2- and 3-photon absorption at $3\omega$, $3\omega/2$, and $\omega$, respectively, are given in Table I for 50 fs FWHM pulses normally incident on WSe$_2$; these values were determined as described in detail earlier [18].

In $n + m$ injection the increased localization of excitations in the Brillouin zone results from interference of the $n$ and $m$ absorption amplitudes, and this increased localization is typically maximized for field strengths giving rise to total probabilities of $n$ and $m$ photon absorption that are nearly equal, $\frac{d}{dt} \langle n_{cv}(k) \rangle_n = \frac{d}{dt} \langle n_{cv}(k) \rangle_m$. For the model we consider, this is the explicit condition for maximum interference between processes, which corresponds to the most localized excitations. In what follows the optical intensities are always set such that this holds. The field amplitudes listed in Table I do not correspond to the condition of maximum interference between any processes; these values specify the upper bound of the perturbative regime for 1-, 2-, or 3-photon absorption processes individually. The values used for plotting are then approximately half of those listed, and scaled appropriately; the strength of the response, namely the number of excited electrons or magnitude of the injected current, depends intimately on the field strength.
FIG. 3: Distribution of injected carriers in crystal momentum space from an optical probe facilitating 1+2 photon absorption; the first Brillouin zone is comprised of a single pair of $\mathbf{K}$ (filled circles) and $\mathbf{K}'$ (empty circles) points.

strengths, however the qualitative features of Brillouin zone densities are independent of these values so long as the interference is maximized.

When considering excitation by incident optical fields, the quantum interference between pathways can be affected by adjusting the frequency, polarization, and phase shift of the fields. As previously mentioned, we consider initially an energy of $\hbar \Omega = 1.5$ eV, and so it is the latter two parameters we initially vary. In what follows we then look at various relative polarizations of the fields, and for each combination of polarizations we vary the phase shift. The quantity we term the “relative phase parameter” arises as the natural parameter to vary; this contains information about phases of both fields. For $1+2$, $2+3$, and $1+3$ absorption processes the relative phase parameters are given by

\begin{align}
\Delta \phi_{12} &= \phi_{2\omega} - 2\phi_{\omega}, \\
\Delta \phi_{23} &= 2\phi_{3\omega/2} - 3\phi_{\omega}, \\
\Delta \phi_{13} &= 3\phi_{3\omega} - 3\phi_{\omega},
\end{align}

respectively, or generally $\Delta \phi_{mn} = n\phi_{\Omega/m} - m\phi_{\Omega/n}$.

A. Co-linearly polarized incident fields

Here the spin of the injected carriers is valley dependent, while the distribution of electronic excitations is valley independent. It is thus sufficient to show the injected carrier distribution about a single valley with the understanding that the same excited charge distribution is present at the other, comprised of electrons of the opposite spin.

1. 1+2 absorption

Yet to gain an overall perspective, in Fig. 3 we plot the relative density of carriers injected in the Brillouin zone for the field at $\omega$ and $2\omega$ both linearly polarized along the $\hat{x}$ direction and $\Delta \phi_{12} = 3\pi/2$, for $2\hbar \omega = 1.5$ eV. In the neighborhood of each valley the carriers are injected with the same polar distribution, indicating that a net current is injected, and that each valley contributes equally to the injected current. In the inset of Fig. 3 we show an enlarged view of the carrier injection about one of the valleys; the width of the arc is associated with the 50 fs pulse duration, and could be made larger or smaller by considering shorter or longer pulses, respectively. In the majority of the following figures we show a view that

FIG. 4: Dependence of $\frac{d}{dt} \langle n_c(k) \rangle_{1+2}$ on $\Delta \phi_{12}$ considering fields of frequency $\omega$ and $2\omega$ both polarized along $\hat{x}$.

(a) $\Delta \phi_{12} = \pi/2$  (b) $\Delta \phi_{12} = 0$  (c) $\Delta \phi_{12} = 3\pi/2$
corresponds to the inset of Fig. 3. Using such a view we show in Fig. 4 how the injected carrier distribution changes with the relative phase parameter $\Delta \phi_{12}$, which controls the interference between the 1- and 2-photon absorption amplitudes.

![Fig. 5: Brillouin zone resolved carrier injection rates arising from single colour absorption, for fields polarized along $\hat{x}$.](image)

In Fig. 5 we show the distribution of carriers injected from (solely) 1-, 2-, or 3-photon absorption, where the different field amplitudes are again chosen so that there is equal total carrier injection from each process. The results here are also valley independent, with carriers of opposite spin being injected about the different valleys. Although each of these carrier distributions is non-polar, they illustrate the general feature, noted above, that the higher-order processes result in more localized regions of injected carriers in the Brillouin zone. We now explain the qualitative differences.

The 1-photon absorption, due solely to interband matrix elements, is peaked in the $\pm \hat{y}$ directions, perpendicular to the direction of the electric field; this follows from (32), for from it we find

$$|v_{c1}(k) \cdot \hat{x}|^2 = \Xi^2 \sin^2 \theta + \frac{\Xi^2 \Delta_{12}^2}{|d_{c1}(k)|^2} \cos^2 \theta,$$

$$|v_{c2}(k) \cdot \hat{y}|^2 = \Xi^2 \cos^2 \theta + \frac{\Xi^2 \Delta_{12}^2}{|d_{c1}(k)|^2} \sin^2 \theta,$$

and thus at larger photon energy, or equivalently at larger $k$, as $\Delta_{12}/|d_{c1}(k)|$ becomes smaller there will be even less injection of carriers near the $\pm \hat{x}$ directions. The 2-photon absorption peaks in the directions $\pm \hat{x}$ associated with the oscillating electric field because of the presence of the intraband matrix element, and exhibits more localization in the Brillouin zone than that of the 1-photon absorption. The strong maxima in the 3-photon absorption are also due to the presence of intraband matrix elements, while the weaker maxima in the $\pm \hat{y}$ directions are due to the terms involving only interband matrix elements. It is clear how the patterns displayed in Fig. 4 result from the interference of the 1- and 2-photon amplitudes responsible for the plots shown in Fig. 5.

2. \textit{2+3 absorption}

In Fig. 6 we plot the carrier injection distributions from 2+3 absorption corresponding to the carrier injection distributions from 1+2 absorption shown in Fig. 4. As the relative phase parameters ($\Delta \phi_{23}$ in the former, $\Delta \phi_{12}$ in the latter) are varied, the two sets of plots show the same qualitative behavior. However, as expected, we see stronger localization of the injected carriers in the 2+3 process than in the 1+2 process. The location of these more localized excitations leads to a larger current injected in 2+3 than in 1+2 absorption at a transition energy of 1.5 eV, as will be discussed in Section VI.

![Fig. 6: Dependence of $\frac{d}{dt} \langle n_c(k) \rangle_{2+3}$ on $\Delta \phi_{23}$ considering fields of frequency $\omega$ and $\frac{3\omega}{2}$ both polarized along $\hat{x}$.](image)

3. \textit{1+3 absorption}

![Fig. 7: Dependence of $\frac{d}{dt} \langle n_c(k) \rangle_{1+3}$ on $\Delta \phi_{13}$ considering fields of frequency $\omega$ and $3\omega$ both polarized along $\hat{x}$.](image)

The injected electronic distributions from 1+3 absorption are qualitatively different than those from 1+2 and 2+3, and are shown in Fig. 7. Here there is no net current injection, but rather a variation in the total density of carriers injected as the relative phase parameter $\Delta \phi_{13}$ is varied. For $\Delta \phi_{13} = \pi/2$, the contributions to $c^{\text{inj}}_{s1+3}(3\omega; k)$ from all $(\tau, s)$ vanish, and at any $k$-point in the Brillouin zone the carriers injected are the sum of those injected at that $k$-point from 1-photon absorption and at 3-photon absorption. There is constructive interference between those two absorption processes at $\Delta \phi_{13} = \pi$, with about 50% more carriers injected than for the same intensities...
at $\Delta \phi_{13} = \pi/2$, and there is destructive interference between the processes at $\Delta \phi_{13} = 0$, with about 50% fewer carriers injected than for the same intensities at $\Delta \phi_{13} = \pi/2$.

### B. Cross-linearly polarized incident fields

The spin of the injected carriers is again valley dependent. However, in contrast to the co-linear case, the injected electronic charge distributions also become valley dependent.

1. 1+2 absorption

In Fig. 8 we plot the distribution of injected carriers for $\hat{e}_{3\omega} = \hat{x}$ and $\hat{e}_{3\omega/2} = \hat{y}$. For $\Delta \phi_{12} = \pi/2$ or $3\pi/2$ the distribution of injected carriers about the different valleys is the same, and exhibits a current in the $-\hat{x}$ direction (for $\Delta \phi_{12} = \pi/2$) or in the $\hat{x}$ direction (for $\Delta \phi_{12} = 3\pi/2$). However, more generally the injected electronic distributions are valley dependent; the most dramatic example is for $\Delta \phi_{12} = \pi$, where currents are injected in the $-\hat{y}$ and $\hat{y}$ directions about $K$ and $K'$ respectively. Here there is no net current injected in either direction. Due to the symmetries of the model, the only non-zero response tensor involved for the net current for the specified polarizations is $n_{1+2}^{\text{xyxy}}$, and so any non-vanishing injected current is in the $\pm \hat{x}$ direction. In fact, for our model, regardless of the crystal axes associated with the cross-linear polarizations, it is always the direction associated with the field facilitating odd number photon absorption (i.e. 1PA, 3PA, etc.) that determines the direction of the net injected current, if there is one. This can be shown analytically using the expressions provided, and is consistent with previously found results for the TMDs [18].

2. 2+3 absorption

![Fig. 8: Dependence of $\frac{d}{d\omega} \langle n_c(k)\rangle_{1+2}$ on $\Delta \phi_{12}$ considering fields orientated according to $\hat{e}_{\omega} = \hat{y}$ and $\hat{e}_{2\omega} = \hat{x}$.](image1)

In Fig. 9 we plot the injected carriers for $\hat{e}_{\omega} = \hat{x}$ and $\hat{e}_{3\omega/2} = \hat{y}$; the corresponding response tensor component is $n_{2+3}^{\text{xyxy}}$. As in the 1+2 case, at $\Delta \phi_{23} = \pi$ there is no net current injected. At $\Delta \phi_{23} = \pi/2$ and $\Delta \phi_{23} = 3\pi/2$ the distribution of injected carriers about the $K$ and $K'$ points are identical, mirroring the situation for 1+2 absorption, and there is a weak current injected in the $-\hat{x}$ direction (for $\Delta \phi_{23} = \pi/2$) and in the $\hat{x}$ direction (for $\Delta \phi_{23} = 3\pi/2$). The carrier distributions injected by cross-linearly polarized incident fields appear to be more strongly localized in the Brillouin zone for 2+3 absorption than for 1+2 absorption, as was seen for co-linearly polarized incident fields. Here, however, it is found that there is a larger current injected from 1+2 absorption than from 2+3 absorption at transition energy of 1.5 eV (see Section VI).

3. 1+3 absorption

![Fig. 9: Dependence of $\frac{d}{d\omega} \langle n_c(k)\rangle_{2+3}$ on $\Delta \phi_{23}$ considering fields orientated according to $\hat{e}_{\omega} = \hat{x}$ and $\hat{e}_{3\omega/2} = \hat{y}$.](image2)

Again, there is no current injection possible for 1+3 absorption. For $\hat{e}_{3\omega} = \hat{x}$ and $\hat{e}_{\omega} = \hat{y}$ the injected carrier distributions are shown in Fig. 10. The distributions about $K$ and $K'$ are the same for $\Delta \phi_{13} = \pi$, but different from each other gener-
by circularly polarized light, and so the single colour absorption parameters are increased. The result of the interference is that one set of regions becomes less populated as the relative phase parameter changes. This parallels what was seen for 1+3 absorption. For a given helicity, say \( \hat{\epsilon}_s \), the injected current varies from 0 to 2 as \( \Delta \phi_{13} \) is varied. Here, however, the injected distributions are valley dependent and the result after summing both valleys is that there is no net injection of carriers; this is consistent with what we display in Fig. 10.

C. Circularly Polarized Incident Optical Fields

Consider first the 1-, 2- and 3-photon absorption processes individually. For a given helicity, say \( \hat{\epsilon}_s \), the distribution of carriers injected by 1-photon absorption will show no dependence on the angle \( \theta \) about the nearby band gap. This follows from (32), which governs the 1-photon absorption rate, and from which we find

\[
\psi_s(k) \cdot \hat{\epsilon}_s = \frac{\varepsilon_0 e^{i (\tau - \tau) \hat{\omega} \cdot \hat{K}}}{\sqrt{2}} \left( \frac{\tau \Delta_s}{|d_s(k)|} + 1 \right),
\]

It is the absolute value squared of (39) that enters in the 1-photon absorption rate for \( \hat{\epsilon}_s \) polarized light, which is independent of \( \theta \). Similar arguments hold for the 2- and 3-photon absorption rates. Thus the localization of injected carriers resulting from a single colour absorption processes using linearly polarized light (recall Fig. 5) is absent for excitation by circularly polarized light, and so the single colour absorption processes do not help in establishing well-localized polar distributions when interference effects are brought into play. Nonetheless, in another sense the use of circular polarizations offers more control compared to the use of linear polarizations, in that continuous variations in the relative phase parameter change the direction of the injected current continuously; this is not the case when exciting electrons using linearly polarized fields.

Notice that near the \( \hat{K} \) point (\( \tau = 1 \)), (39) will be larger than about the \( \hat{K}' \) (\( \tau = -1 \); indeed, as the excitation energy decreases to the band gap energy and \( |d_s(k)| \to \Delta_s \), there will be no carriers injected about \( \hat{K}' \) by light of this helicity.

1. Equal helicities

We first consider 1+2 absorption, with \( \hat{\epsilon}_s \) being the polarization for the fields at both frequencies. The injected carrier distributions about the \( \hat{K} \) point are shown in Fig. 11; those about \( \hat{K}' \) are qualitatively the same, but with far fewer carriers injected. As the relative phase parameter \( \Delta \phi_{12} \) is varied from 0 to 2\( \pi \) the direction of the injected current varies continuously over the same angular range in real space. Since more carriers are injected about the \( \hat{K} \) valley than about the \( \hat{K}' \) valley, and as carriers injected in different valleys have different spins, the injected current will be spin polarized. This was discussed previously [18]. The scenario for 2+3 absorption is qualitatively the same and thus not included as a figure; as \( \Delta \phi_{23} \) varies from 0 to 2\( \pi \) the direction of the injected spin current varies from 0 to 2\( \pi \) in real space.

![Fig. 11: Dependence of \( \frac{d}{d\phi} \langle n_s(k) \rangle \) on \( \Delta \phi_{12} \) considering fields of frequency \( \omega \) and 2\( \omega \) both circularly polarized in \( \hat{\epsilon}_s \).](image)

For 1+3 absorption there is no injected current, as expected, but there is a non-uniform distribution of injected carriers in the Brillouin zone that rotates as the relative phase parameter \( \Delta \phi_{13} \) varies; this is shown in Fig. 12. Here the total number of carriers injected does not vary with \( \Delta \phi_{13} \), as was possible for linearly polarized excitation, but since there are more carriers injected around \( \hat{K} \) than \( \hat{K}' \), and since the spin of the injected carriers is correlated with the valley, the injected carriers are spin polarized.

The scenario for \( \hat{\epsilon}_s \) polarized light is essentially the same
for all interference processes, but with the predominant valley and spin of the injected carriers reversed.

2. Opposite Helicities

For a transition precisely at the band gap facilitated by fields having opposite helicity, carriers are injected into each valley by only one of the two frequencies, namely the frequency associated with the helicity of light that couples to that particular valley, and of course there is no interference. For higher excitation energies interference does arise because each polarization injects carriers into both valleys, although more into one than into the other. Yet in our model for the TMDs there is no variation in the number of carriers injected as the appropriate phase parameter is varied, nor is there net current injected for any of the interference processes. However, anisotropic carrier distributions are injected that rotate as with variation in the relative phase parameter. For 1+2 absorption the anisotropic distribution has three-fold rotational symmetry, for 1+3 absorption it has four-fold rotational symmetry, and for 2+3 interference it has five-fold symmetry. We show the first of these for our excitation energy of $h\Omega = 1.5$ eV in Fig. 13.

![Fig. 12: Dependence of $\Delta \phi_{13}$ on $\Delta \phi_{13}$ considering fields of frequency $\omega$ and $3\omega$ both circularly polarized in $\hat{e}_z$.](image)

![Fig. 13: Dependence of $\Delta \phi_{12}$ on $\Delta \phi_{12}$ considering fields oriented according to $\hat{e}_{\omega'} = \hat{e}_+ \text{ and } \hat{e}_{2\omega'} = \hat{e}_-$.](image)

VI. FREQUENCY DEPENDENCE OF INJECTION COEFFICIENTS

As the excitation energy $h\Omega$ is varied, the carrier and current injection distributions shown above will change. For an overview of this we look at plots of the injection coefficients themselves.

A. Carrier injection coefficients

In Fig. 14 we show contributions to the carrier injection rate about the $K$ point arising solely from single colour absorption: $\xi_1$, $\xi_2$, and $\xi_3$. The total coefficient, $\xi_n$, is given as a sum over contributions $\xi_{n,\tau s}$ associated with valley $\tau$ and spin $s$, $\xi_n = \sum_{\tau s} \xi_{n,\tau s}$; however, to label the plots we use the notation $\xi_{n,\tau} = \sum_s \xi_{n,\tau s}$. In Fig. 15 we display the interference terms that give rise to a non-vanishing carrier injection rate. As it happens, this term is only non-vanishing in the case of 1 + 3 absorption.

We find the real valued components of the response tensor to be valley independent, while the imaginary parts differ by a sign between $K$ and $K'$. So the real part characterizes the total carrier injection, while the imaginary part characterizes the imbalance of injected carriers between the valleys. For 1-photon absorption we have $\xi_{\text{xx}} = \xi_{\text{yy}}$ and they are both real; here and below we only show independent components. The cross term $\xi_{\text{xy}}$ is imaginary, reflecting the structure of the interband matrix element (32), and close but not identically equal in magnitude to $\xi_{\text{yx}}$. There is the usual step-like increase in the 1-photon absorption coefficient at the band gap because the matrix element (32) is finite there, and a second step-like increase in magnitude at the onset of absorption from the lower valence band.

Unlike the 1-photon carrier injection coefficients, the 2-photon injection coefficients have a smooth initial onset because of the presence of intraband matrix elements (33) appearing in the expression (34) for the $R_{\text{cv}}^{(2\text{ab})}$, which arises in the expression (20) for $\xi_2$: these intraband elements vanish at the band gap and change continuously as one moves away from it. The onset of absorption from the lower valence band is also smooth for the same reason. The overall magnitudes of the $\xi_2$ coefficients drop off faster with increasing excitation energy than does that of the $\xi_1$ coefficients because a larger number of frequencies appear in the denominator of the coefficients (7) $R_{\text{cv}}^{(N)}(k)$ as $N$ is increased.

The 3-photon carrier injection coefficients $\xi_3$ share some of the features of the $\xi_1$ and some of the $\xi_2$, since $R_{\text{cv}}^{(3)}(k)$ contains pure interband (PR) contributions (as does $R_{\text{cv}}^{(1)}(k)$) and interband-intraband contributions (RA) that involve both types of matrix elements (as does $R_{\text{cv}}^{(2)}(k)$); this also leads to
the coefficients $\xi_3$ exhibiting a more complicated energy dependence than the elements of either $\xi_1$ or $\xi_2$. For example, the sharp onset at the band edge of the different components of $\xi_3$ is due to the PR contribution, while the later, smoother rise (or fall) in the different components is due to an increasing contribution from the RA contributions as the magnitude of $\mathbf{v}_{cc}(k) - \mathbf{v}_{vs}(k)$ increases at larger $k$; certain components of $\xi_3$ can actually vanish as the PR and RA contributions cancel. The interference coefficient $\xi_{1+3}$ again shows the presence of PR and RA contributions. The first leads to the step-like increase at the band edge, and another step-like change at the onset of absorption from the lower valence band. The second leads to the smoother change with increasing excitation energy that actually leads to a change in sign of the non-
vanishing coefficients as the PR and RA contributions cancel one another. Note that all components of \( \Re(\xi_{1+3}) \), which govern the net carrier injection, vanish at a particular excitation energy \( \hbar \Omega = 1.31 \) eV. Thus the destructive interference between the PR and RA contributions (see (34) for \( K_{c2}^{3ab} \)) leads to a frequency region where there is very small coherent control of the carrier injection rate. At the energy that \( \Re(\xi_{1+3}) \) vanishes \( \Im(\xi_{1+3}) \) is not strictly vanishing, and therefore there will be carrier injection interference in both the \( K \) and \( K' \) valleys, but the interference effects will cancel when producing the total carrier injection rate. Nonetheless, at this energy \( \Im(\xi_{1+3}) \) is very small, and so even the “valley-by-valley” interference will be very small.

**B. Current injection coefficients**

The contributions to the current injection coefficients \( \eta_{1+2} \) and \( \eta_{2+3} \) from the \( K \) valley are shown in Fig. 16 and 17, respectively, as a function of excitation energy; an analogous notation to that used for plotting carrier injection rates is adopted here. In contrast to the carrier injection, here we find the imaginary valued components of the response tensor to be valley independent, while the real parts differ by a sign between \( K \) and \( K' \), so it is the imaginary parts that completely characterize the charge current injected into the system. Earlier work [24] on simpler systems showed that including the Coulomb interaction between injected electrons and holes led to the prediction of a phase shift introduced in the response, and thus to an expected maximum injection current occurring at a relative phase parameter different that \( \Delta \phi_{12} \) (or \( \Delta \phi_{23} \)) = \( \pi/2 \) and \( 3\pi/2 \). That should be expected here as well; we plan to investigate the inclusion of this effect in a later publication.

Neither \( \eta_{1+2} \) nor \( \eta_{2+3} \) show step-like behavior as the energy crosses band gaps because of the involvement of intra-band matrix elements in each term of the expression, again arising through the \( K_{c2}^{3ab} \) term. Likewise there is no step-like behavior at the onset of the absorption from the lower valence.

**FIG. 16:** Excitation energy dependence of the independent components of the current injection response tensor for 1+2 photon absorption processes. We plot components about a single valley, \( K \), and do not show vanishing components.

**FIG. 17:** Excitation energy dependence of the independent components of the current injection response tensor for 2+3 photon absorption processes. We plot components about a single valley, \( K \), and do not show vanishing components.
band. Again \( \eta_{2+3} \) is suppressed more at excitation energies greater than \( \eta_{1+2} \) because of frequency factors in the denominators. The nature of the excitation energy dependence of \( \eta_{2+3} \) is much more complicated than that of \( \eta_{1+2} \), and arises again primarily because of the combination of PR and RA terms in the third order response. Note that the sign of many of the imaginary components of \( \eta_{2+3} \) changes as a function of excitation energy, exhibiting the interplay between those two terms in the third order response. Since it is these components that characterize the total current injection in 2+3 absorption, we can expect interesting consequences in the excitation energy dependence of the injected current, which we consider next.

C. Swarm Velocities

We now characterize the average velocity of the injected carriers by considering the “swarm velocity”, which is the current injection rate divided by the total charge injection rate \[31]. For 1+2 absorption it is given by

\[
v_{\text{swarm}}(\hbar\Omega) = \frac{\frac{\partial}{\partial t} \langle J \rangle_{1+2}}{e \left( \frac{d}{dt} \langle n_c \rangle_1 + \frac{d}{dt} \langle n_c \rangle_{1+2} + \frac{d}{dt} \langle n_c \rangle_2 \right)} \bigg|_{\text{max}}^\Omega, \quad (40)
\]

and for 2+3 absorption it is given by

\[
v_{\text{swarm}}(\hbar\Omega) = \frac{\frac{\partial}{\partial t} \langle J \rangle_{2+3}}{e \left( \frac{d}{dt} \langle n_c \rangle_2 + \frac{d}{dt} \langle n_c \rangle_{2+3} + \frac{d}{dt} \langle n_c \rangle_3 \right)} \bigg|_{\text{max}}^\Omega, \quad (41)
\]

where the subscript \( \text{max} \) indicates that the relative amplitudes of the fields appearing, and the relative phase parameter (\( \Delta \phi_{12} \) and \( \Delta \phi_{23} \) respectively), are set to guarantee that the magnitude of the swarm velocity is a maximum. In both 1+2 and 2+3 excitation the phase parameter that does this can be \( \Delta \phi = \pi/2 \) or \( \Delta \phi = 3\pi/2 \), and we choose the latter.

We look at the examples of co- and cross-linearly polarized light for the excitation scenarios, where the current injected is in the \( \pm \hat{x} \) direction. Recall that in the co-linear case this arises with all fields polarized in the \( \hat{x} \) direction, while in the cross-linear case it arises for \( \hat{e}_{3\omega} = \hat{x} \) and \( \hat{e}_{3\omega/2} = \hat{y} \) in 1+2 absorption, and for \( \hat{e}_{\omega} = \hat{x} \) and \( \hat{e}_{3\omega/2} = \hat{y} \) in 2+3 absorption. As the excitation energy is increased we expect the magnitude of the swarm velocity to increase, simply because the carriers injected will have larger velocities.

Comparing two swarm velocities at the same excitation energy gives a measure of how well localized the carriers are in the Brillouin zone. Under the specified excitation conditions we plot the swarm velocities for excitations facilitated by co-linearly polarized light in Fig. 18a, and by cross-linearly polarized light in Fig. 18b. The former are generally larger than the latter, and in particular the latter are very small near an excitation energy of 1.5 eV, especially for 2+3 absorption as noted in our discussion above; these velocities do increase at larger excitation energy. In general the swarm velocities for 2+3 absorption become larger than those of 1+2 absorption for high enough excitation energy. At low excitation energies we see a reversal of the swarm velocity for 2+3 absorption with increasing excitation energy, again due to interplay between the PR and RA contributions to the 3-photon absorption amplitude, and as a result its magnitude is smaller than the swarm velocity for 1+2 absorption. The PR contribution to the amplitude is finite at the band gap, but the RA contribution vanishes. As the excitation energy moves away from the gap, the RA contribution becomes finite with net opposite sign of the PR term; at an excitation energy of \( \hbar\Omega = 1.24 \text{ eV} \) for
co-linearly polarized excitation, and \( h \Omega = 1.29 \) eV for co-
linearly polarized light, the contributions cancel, leading to
no net current injection. Within the model adopted here, this
adds another level of control over the injected current, above
and beyond what can be done by adjusting field intensities and
the relative phases parameters, and only arises for 2+3 absorp-
tion. Also, all the swarm velocity plots show a discontinuous
jump as the energy crosses the second band gap, as would be
expected from the behavior of the injection coefficients.

VII. RESULTS & DISCUSSION

We have shown that the quantum interference arising from
2+3 photon absorption can give rise to significantly more lo-
calized distributions of electronic excitations in the Brillouin
zone than the 1+2 counterpart (see Fig. 18). The primary
reason for this is the increased number of intraband velocity
matrix elements in the transition coefficients at third or-
der perturbation theory. The increased localization of these
distributions is most apparent for co-linearly polarized inci-
dent optical fields, and it is also this orientation of fields that
lends itself most to the idea of using QuIC as "tweezers in
the Brillouin zone" (Fig. 6); using quantum interference of
excitation processes as a mechanism to place carriers where
the Brillouin zone would normally be.

Also, all the swarm velocity plots show a discontinuous
jump as the energy crosses the second band gap, as would be
expected from the behavior of the injection coefficients.

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Appendix A: Independent Response Tensor Components

1. Carrier Injection Rate:

The non-vanishing, independent, valley- and spin-
dependent response tensor components corresponding to the
carrier injection rate are given for the indicated photon ab-
sorption processes. The relation of these components to the
carrier injection rate is given in Section III, and as there we
use the notation \( \xi_n = \sum_{rs} \xi_{n;rs} \).

1 Photon Absorption:

\[
\begin{align*}
\xi_{1;rs}^{xx}(2\omega) &= \frac{\Theta(2\nu-2\Delta_\nu)e^2}{16\hbar^2 \omega} \left( 1 + \frac{\Delta_\nu}{\omega} \right) \\
\xi_{1;rs}^{yy}(2\omega) &= -i \frac{\Theta(2\nu-2\Delta_\nu)e^2}{8\hbar^2 \omega} \left( 1 + \frac{\Delta_\nu}{\omega} \right) 
\end{align*}
\]

(A1)

2 Photon Absorption:

\[
\begin{align*}
\xi_{2;rs}^{xxx}(2\omega) &= \frac{\Theta(2\nu-2\Delta_\nu)e^4e^2}{16\hbar^2 \omega^2} \left( 1 + \frac{\Delta_\nu}{\omega} \right) \left( 1 + \frac{\Delta_\nu}{2\omega} \right) \\
\xi_{2;rs}^{xyy}(2\omega) &= -i \frac{\Theta(2\nu-2\Delta_\nu)e^4e^2}{32\hbar^2 \omega^2} \left( 1 + \frac{\Delta_\nu}{\omega} \right) \left( 1 + \frac{\Delta_\nu}{\omega} \right) \\
\xi_{2;rs}^{yxy}(2\omega) &= \frac{\Theta(2\nu-2\Delta_\nu)e^4e^2}{48\hbar^2 \omega^2} \left( 1 + \frac{\Delta_\nu}{\omega} \right) \left( 1 + \frac{\Delta_\nu}{\omega} \right) \\
\xi_{2;rs}^{yyx}(2\omega) &= -i \frac{\Theta(2\nu-2\Delta_\nu)e^4e^2}{48\hbar^2 \omega^2} \left( 1 + \frac{\Delta_\nu}{\omega} \right) \left( 1 + \frac{\Delta_\nu}{\omega} \right) 
\end{align*}
\]

(A2)

1+3 Absorption:

\[
\begin{align*}
\xi_{1+3;rs}^{xxx}(3\omega) &= -i \frac{\Theta(3\nu-2\Delta_\nu)e^2}{18\hbar^2 \omega} \left( 2 + \frac{1}{2} \right) \frac{\Xi^2 k_{rs}^2}{k_{rs}} - \frac{9}{8} \frac{\omega_2^2}{\xi^4 k_{rs}^2} \left( 2 + \frac{1}{2} \right) \frac{\Xi^2 k_{rs}^2}{k_{rs}} - \frac{9}{8} \omega_2^2 \\
\xi_{1+3;rs}^{xyy}(3\omega) &= -i \frac{\Theta(3\nu-2\Delta_\nu)e^2}{18\hbar^2 \omega} \left( 1 + \frac{2}{3} \right) \frac{\Xi^2 k_{rs}^2}{k_{rs}} - \frac{9}{8} \omega_2^2 \\
\xi_{1+3;rs}^{yxy}(3\omega) &= i \frac{\Theta(3\nu-2\Delta_\nu)e^2}{18\hbar^2 \omega} \left( 2 + \frac{1}{2} \right) \frac{\Xi^2 k_{rs}^2}{k_{rs}} - \frac{9}{8} \omega_2^2 \\
\xi_{1+3;rs}^{yyx}(3\omega) &= -i \frac{\Theta(3\nu-2\Delta_\nu)e^2}{18\hbar^2 \omega} \left( 1 + \frac{2}{3} \right) \frac{\Xi^2 k_{rs}^2}{k_{rs}} - \frac{9}{8} \omega_2^2 
\end{align*}
\]

(A3)
3 Photon Absorption:

\[
\begin{align*}
\xi_{3,\tau,\tau}^{XXX}(3\omega) &= \frac{\theta}{3!} \frac{\Theta(3\omega-2\Delta_m)}{2\Delta_m} \left( 4 \pi^6 - \pi^4 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 9 \frac{3}{8} \right) k_{\tau}^2 - \left( \frac{2\pi^2}{\Delta_m} \right)^6 \left( 20 + \frac{1}{2} \right) \frac{1}{4} k_{\tau}^6 \right) \\
\xi_{3,\tau,\tau}^{XYXY}(3\omega) &= \frac{\theta}{3!} \frac{\Theta(3\omega-2\Delta_m)}{2\Delta_m} \left( 4 \pi^6 - \pi^4 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 9 \frac{3}{8} \right) k_{\tau}^2 - \left( \frac{2\pi^2}{\Delta_m} \right)^6 \left( 20 + \frac{1}{2} \right) \frac{1}{4} k_{\tau}^6 \right) \left( 3 + \frac{3}{8} \right) + \frac{3}{4} \pi^6 \right) \\
\xi_{3,\tau,\tau}^{XYXY}(3\omega) &= \frac{\theta}{3!} \frac{\Theta(3\omega-2\Delta_m)}{2\Delta_m} \left( 4 \pi^6 - \pi^4 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 9 \frac{3}{8} \right) k_{\tau}^2 - \left( \frac{2\pi^2}{\Delta_m} \right)^6 \left( 20 + \frac{1}{2} \right) \frac{1}{4} k_{\tau}^6 \right) \left( 5 + \frac{11}{32} \right) \frac{1}{4} k_{\tau}^4 \right) \\
\xi_{3,\tau,\tau}^{XYXY}(3\omega) &= \frac{\theta}{3!} \frac{\Theta(3\omega-2\Delta_m)}{2\Delta_m} \left( 4 \pi^6 - \pi^4 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 9 \frac{3}{8} \right) k_{\tau}^2 - \left( \frac{2\pi^2}{\Delta_m} \right)^6 \left( 20 + \frac{1}{2} \right) \frac{1}{4} k_{\tau}^6 \right) \left( 9 \frac{3}{8} \right) \frac{2}{4} k_{\tau}^4 \right) \\
\xi_{3,\tau,\tau}^{XYXY}(3\omega) &= \frac{\theta}{3!} \frac{\Theta(3\omega-2\Delta_m)}{2\Delta_m} \left( 4 \pi^6 - \pi^4 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 9 \frac{3}{8} \right) k_{\tau}^2 - \left( \frac{2\pi^2}{\Delta_m} \right)^6 \left( 20 + \frac{1}{2} \right) \frac{1}{4} k_{\tau}^6 \right) \left( 2 \right) \frac{1}{4} k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 + \pi^2 \left( \frac{2\pi^2}{\Delta_m} \right)^2 \left( 2 + \frac{3}{8} \right) k_{\tau}^2 \right) \left( 8 + \frac{1}{4} \right) \frac{1}{4} k_{\tau}^4 \right) \\
\end{align*}
\]

(A4)

2. Current Injection Rate:

The non-vanishing, independent, valley- and spin-dependent response tensor components corresponding to the current injection rate are given for the indicated photon absorption processes. The relation of these components to the current injection rate is given in Section III, and as there we use the notation \( n_n = \sum_{\tau_n} n_{n,\tau_n} \).

1+2 Absorption:

\[
\begin{align*}
\eta_{1,2,\tau}^{XXX}(2\omega) &= \frac{i \theta(2\omega-2\Delta_m)\delta^2}{h \omega} \left( 1 - \frac{\lambda_1^2}{\omega} \right) \left( 1 + \frac{\lambda_1^2}{\omega} \right) \\
\eta_{1,2,\tau}^{XYXY}(2\omega) &= \frac{i \theta(2\omega-2\Delta_m)\delta^2}{h \omega} \left( 1 - \frac{\lambda_1^2}{\omega} \right) \\
\eta_{1,2,\tau}^{XYXY}(2\omega) &= -\frac{i \theta(2\omega-2\Delta_m)\delta^2}{h \omega} \left( 1 - \frac{\lambda_1^2}{\omega} \right) \\
\eta_{1,2,\tau}^{XYXY}(2\omega) &= \frac{i \theta(2\omega-2\Delta_m)\delta^2}{h \omega} \left( 1 - \frac{\lambda_1^2}{\omega} \right) \\
\eta_{1,2,\tau}^{XYXY}(2\omega) &= 0 \\
\end{align*}
\]

(A5)

2+3 Absorption:

\[
\begin{align*}
\eta_{2,3,\tau}^{XXX}(3\omega) &= \frac{i \theta(3\omega-2\Delta_m)\delta^3}{h \omega} \left( 1 - \frac{\lambda_2^2}{\omega} \right) \left( 1 - \frac{\lambda_2^2}{\omega} \right) \left( 1 - \frac{\lambda_2^2}{\omega} \right) \\
\eta_{2,3,\tau}^{XYXY}(3\omega) &= \frac{i \theta(3\omega-2\Delta_m)\delta^3}{h \omega} \left( 1 - \frac{\lambda_2^2}{\omega} \right) \\
\eta_{2,3,\tau}^{XYXY}(3\omega) &= \frac{i \theta(3\omega-2\Delta_m)\delta^3}{h \omega} \left( 1 - \frac{\lambda_2^2}{\omega} \right) \\
\eta_{2,3,\tau}^{XYXY}(3\omega) &= \frac{i \theta(3\omega-2\Delta_m)\delta^3}{h \omega} \left( 1 - \frac{\lambda_2^2}{\omega} \right) \\
\eta_{2,3,\tau}^{XYXY}(3\omega) &= 0 \\
\end{align*}
\]

(A6)

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[31] Notice that for constant injection rates, the expression for the swarm velocity reduces to $v_{swarm}(\hbar \Omega) = \langle J \rangle / e \langle n_c \rangle$. 
