Nonvanishing sub-gap photocurrent as a probe of correlation effects

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For semiconductors and insulators, it is commonly believed that in-gap transitions into non-localized states are smoothly suppressed in the clean limit, i.e. at zero temperature they vanish even without accounting for localization effects. We show in a counterexample that this intuition does not generalize beyond the linear response. Namely, we find that the dc current due to the bulk photovoltaic effect can be finite and mostly temperature independent in an allowed window of sub-gap transitions. As the reason we identify a nontrivial phase accumulation which is related to the fact that the electron motion due to the applied electric field is, within limits, coherent. We expect that a moderate range of excitation energies, where this effect is observable, lies between the bulk energy gap and the mobility edge. We find that the sub-gap dc-current is temperature-independent for non-interacting systems but temperature-dependent for strongly interacting systems. Thus, the sub-gap response may be used to distinguish whether a state is single-particle localized or many-body localized.

Introduction. — The electronic motion in periodic systems with low electron density is sensitively dependent on the interplay of disorder and interactions. In the absence of interactions, electronic response can be formulated in the language of Anderson localization by assigning a localization length to each momentum eigenstate [1, 2]. The transition from infinite to finite localization length then immediately defines a mobility edge differentiating wave-functions which do contribute to transport from those who do not. Most importantly, a decreasing density of states (DOS) is accompanied by a mobility edge separating delocalized states at high DOS from localized ones at low DOS [3].

For sub-gap linear optical response of clean systems with a direct band gap \( \Delta \), these localization effects are usually not important. Namely, the ac-conductivity \( \sigma^{\alpha\alpha}(\omega) \), associated with a current \( j^{\alpha}(\omega) = \sigma^{\alpha\alpha}(\omega)E_{\alpha}(\omega) \) is given by the Kubo formula. Therefore, for \( \omega < \Delta \), the conductivity \( \sigma^{\alpha\alpha}(\omega) \) decreases smoothly as the density of states vanishes inside the bandgap. This behavior is captured by assigning to eigenstates of energy \( \varepsilon_m(k) \) with band index \( m \) a finite linewidth \( (2\gamma)^{-1} \) through the so-called adiabatic switching \( \omega \to \omega + i \gamma \) in the inverse propagator \( G_0^{-1}(\omega, k) = \omega + i0^+ - \varepsilon_m(k) \). At small temperatures, this procedure will induce a finite DOS of size \( O(\gamma) \) in the gap of fully gapped insulators and semiconductors due to Lorentz broadening of the band energies, which vanishes smoothly as \( \gamma \to 0 \). The same holds true for the conductivity \( \sigma^{\alpha\alpha}(\omega) \).

Also in the theory of non-linear optical response current creation is discussed without resorting to localization [4–9]. It is a widely held belief that one can smoothly recover a vanishing non-linear conductivity in insulators and semiconductors upon taking the limit \( \gamma \to 0 \), just like at linear order. Indeed, this has to be the case if all processes are adiabatic [10].

However, non-linear optical response is related to the joint density of states of the valence band and conduction band, which can remain finite at in-gap energies even in the clean limit. As we show in this letter, under certain circumstances it is therefore possible to exploit sub-gap optical response to investigate the interplay of disorder and interactions asymptotically for \( \gamma \to 0 \), opening a new avenue to explore the physics of weak localization and also many-body localization [11, 12] in condensed matter systems.

In the following, the current is driven by an extrinsic field-induced polarization as it typically arises in second-order optical response [5, 13–23]. To this end, we study the dc-conductivity \( \sigma^{\alpha\alpha}_n(0; \omega, -\omega) \) using the second-order perturbative expansion in the veloc-
ity gauge, defined through a dc-current density \(j'(0) = \sigma^{abc}(0; \omega, -\omega)E_a^*(\omega)E_b(\omega)\) (cf. Fig. 1). We consider an insulator with a gap \(\Delta\), whose states have a finite linewidth for intra- and interband processes, denoted \(\gamma\) and \(\Gamma\). By expanding the expressions in the regime where \(\omega < \Delta\), we find systems with time-reversal symmetry (TRS) a suppression of the sub-gap conductivity \(\sigma_{\text{sub}}(\omega) = \sigma^{a,a}(\omega < \Delta)\) in accordance with the expected scaling in the regime \(\sigma_{\text{sub}} \sim \Gamma\), which vanishes smoothly in the clean limit. However, for systems lacking TRS the conductivity turns out to be non-vanishing as a function of \(\Gamma, \gamma \rightarrow 0\) where \(\gamma/\Gamma\) fixed. As a reason we note that the non-linear optical conductivity is the result of several different two- and three-band processes which normally interfere destructively, but which do no longer annihilate each other completely once TRS is broken. In optical response, such an interference between different processes is possible because the interaction with the electric field is, at least within some bounds, coherent. In the language of second-order response theory, we observe that in the sub-gap regime the injection current equals the negative of the shift current only for TRS-preserving materials. We emphasize that the lack of cancellation in the case of light, the sequential two-photon processes are up to order \(O(\frac{\omega}{\Delta})\) given by

\[
\eta_1 = 4C \text{Re}\left[\frac{1}{i\omega} \int_\mathbb{R} \int f_{nm} \frac{\epsilon_{nm}^a|^2 v_{nm}^a}{\epsilon_{nm} + i\Gamma}\right],
\]

where \(C = -\alpha e^3/\omega^2 h^2\) and \(\alpha\) the ratio of transport lifetime to quasiparticle lifetime [25]. When TRS is present, \(T\eta_1T^{-1} = -\eta_1\), and as expected this so-called injection current vanishes identically. However, when TRS is absent, \(\eta_1\) can take a non-zero value. By using the replacement \(v_{nm}^a = i\epsilon_{nm}^a r_{nm}^a\), where \(n \neq m\), we obtain \(\eta_1 = -4C \sum_{nm} f_{nm} \int \frac{\Gamma}{\omega} |r_{nm}^a|^2 v_{nn}^a\), after explicitly letting \(\Gamma \rightarrow 0\). The remaining terms with photon-dressed current vertices can be written as

\[
\eta_2 = 2C \text{Re}\left[\frac{1}{\omega} \int \sum_{nm} f_{nm} w_{nm}^a v_{mn}^a\right].
\]

We used the fact that \(w_{nm}^a = \partial v_{nm}^a + i[v_{nm}^a, r_{nm}^a]_{nm}\). The leading order component is then \(\eta_2 = 2C \sum_{nm} f_{nm} |r_{nm}^a|^2 v_{nn}^a\), where we considered only the real part of the numerator in expanding \(\eta_2\) as the imaginary part is subleading. The total response \(\sigma_{\text{sub}} = \eta_1 + \eta_2\) therefore becomes

\[
\sigma_{\text{sub}} = -2C \left(\frac{2\pi}{\gamma} - 1\right) \left(\int \sum_{nm} f_{nm} |r_{nm}^a|^2 v_{nn}^a\right).
\]

Again, \(\sigma_{\text{sub}}\) is odd under TRS, and hence vanishes identically under an applied linearly polarized field. We therefore conclude that the T-symmetric system has a sub-gap dc-conductivity no larger than \(\sigma_{\text{sub}} \sim \frac{\Gamma}{\Gamma}\). Note that in this case, the smooth limit \(\Gamma \rightarrow 0\) exists, meaning that \(\sigma_{\text{sub}} \rightarrow 0\) independently of the precise finite value of the ratio \(\gamma/\Gamma\). The main result of this letter is contained in Eq. (3), which is the leading order term for a system lacking TRS. In the conventional language of non-linear optical response, Eq. (3) is the sum of the injection current \(\eta_1\) and the shift current \(\eta_2\). The factor of \(\frac{2\pi}{\gamma} - 1\) appearing in this relation implies that for the value of \(\gamma/\Gamma = 2\) the injection and shift currents cancel below the gap, up to regular terms of size \(\frac{\Gamma}{\Gamma}\). For any other value of \(\gamma/\Gamma\) it remains finite. Since the dc-current is a function of the dimensionless parameter \(\gamma/\Gamma\), it is insensitive to the absolute values of the broadening induced by \(\gamma, \Gamma\). Most notably, the clean limit of \(\gamma, \Gamma \rightarrow 0\) with \(\gamma/\Gamma\) fixed does not lead to a smoothly vanishing dc-current according to Eq. (3). By no means does this implies that the current persists at any frequency below the gap, as it does not consider the effects of localization which will inevitably cut off such a behavior at a small density of states. However, the result does not require a specific microscopic model for the type of broadening which leads to the finite relaxation rates \(\gamma, \Gamma\), except to require that these rates can be approximated by some relaxation time approximation. We propose to make use of this desirable property as a probe of localization physics.
Example.— For demonstration, we consider a 2d Haldane model with next-nearest neighbor spin-orbit-like coupling, similar to the form suggested by Kane and Mele. In an infinite 2d-system all eigenstates are localized, meaning that there is no mobility edge. However, it is still sensible to discuss currents and localization effects for mesoscopic systems with a size smaller than the largest localization length [26]. Our Hamiltonian reads,

$$H = t_1 \sigma_x + t_2 \sigma_y + m_1 \sigma_z s_z + m_2 \sigma_z + \delta(k)\sigma_z s_z.$$  \hspace{1cm} (4)

Where $t_1 = \text{Re}[t(k)]$, $t_2 = \text{Im}[t(k)]$, such that

$$t(k) = -t\left(e^{i(\frac{3}{2}k_x+k_y)} + e^{i(-\frac{3}{2}k_x+k_y)} + e^{-ik_y}\right).$$

The nearest-neighbor distance of the honeycomb lattice is $b = 1$, $\sigma$ and $s$ relate to the sub-lattice and spin degrees of freedom, respectively. Inversion in this system takes the form $P = \sigma_z K$, where $K : k \rightarrow -k$, and time-reversal is $T = is_y C K$, $C$ corresponds to complex conjugation. We set $m_1 \neq 0$, $m_2 = 0$ for TRS-breaking, and $m_1 = 0$, $m_2 \neq 0$ for the T-symmetric case. The spin-orbit coupling term has the form,

$$\delta(k) = -td\left[\sin\left(\sqrt{3}k_x\right) - \sin\left(\sqrt{3}k_x/2 - 3k_y/2\right) - \sin\left(\sqrt{3}k_x/2 + 3k_y/2\right)\right].$$ \hspace{1cm} (5)

Fig. 2 shows the results for the conductivity $\sigma^{xz,xy} = -\sigma^{yz,yz}$, for linearly polarized light using $m_1 \neq 0$.

Crystal symmetries dictate that $\sigma^{xx,xx} = -\sigma^{yy,yy} = -\sigma^{yx,yx}$, generically, while $\sigma^{yy,yy} = 0$, and hence $\sigma^{xx,xx} = \sigma^{yy,yx} = \sigma^{yx,xx} = 0$. Several different ratios $\gamma/\Gamma$ are shown, with the gap presented as the dashed line. We find that for any ratio except $\gamma/\Gamma = 2$, the conductivity below the gap quickly converges to a non-zero value that depends on the ratio $\gamma/\Gamma$. In particular, for values $\gamma/\Gamma > 2$, we find that the conductivity changes sign as the frequency crosses the gap. It is only for the ratio $\gamma/\Gamma = 2$ that we observe a scaling as $\sim \Gamma^2$, in agreement with Eq. (3), it is this ratio that the leading order term $\eta_1 + \eta_2$ disappears and only sub-leading terms remain. In conclusion, the subgap response is markedly different between $\gamma/\Gamma = 2$ and $\gamma/\Gamma \neq 2$, with asymptotics $\sigma^{aa,aa} \sim \mathcal{O}(\omega^{-2})$ and $\sigma^{aa,ai} \sim \mathcal{O}(\omega^{-4}\Gamma^2)$ respectively. In Fig. 3, the conductivity component $\sigma^{yy,yy} = -\sigma^{xx,xy}$ is shown for $m_2 \neq 0$, which is the only one present due to mirror symmetries and TRS. Due to TRS, we recover the expected result that $\sigma \sim \Gamma$ below the gap. Notably, the conductivity can smoothly be continued to $\Gamma \rightarrow 0$, and the result is independent of the ratio $\gamma/\Gamma$. For values above the gap, Fig. 2 shows the appearance of the injection current, which scales as $\frac{1}{\tau}$, while in Fig. 3 we see the shift current emerging, which for $\Delta \ll \omega$ is independent of $\Gamma$. Fig. 4, shows the comparison between $\eta_1 + \eta_2$ and the complete second order response as a function of the ratio $\gamma/\Gamma$. The close agreement confirms that the value of the sub-gap conductivity is dominated by this term. As has been noted in [27], the ratio of $\gamma/\Gamma = 2$ is the result of assuming a particular relaxation mechanism for the different bands. This condition corresponds to the replacement in the response of $\omega \rightarrow \omega + i\Gamma$, such that in denominators with two frequencies one should put $\omega + \omega_l \rightarrow \omega_1 + \omega_2 + 2i\Gamma$. However, this prescription is not always justified. Namely, the ratio $\gamma/\Gamma$ can be written using the lifetimes $\tau_0$ and $\tau_1$ of the valence band and the conduction band as

$$\frac{\gamma}{\Gamma} = \frac{1}{\frac{1}{\tau_0} + \frac{1}{\tau_1}}.$$ \hspace{1cm} (6)

In order to fulfill the condition $\gamma/\Gamma \sim 2$ which enforces the cancellation in the TRS-broken case, it must thus hold that $\tau_1/\tau_0 \rightarrow \infty$. In other words, the quasiparticle in the empty conduction band has a lifetime greatly exceeding the ground state quasiparticle lifetime. There is essentially only one situation in which this seems reasonable, which is for a strongly-correlated ground state.

We note that ratios such that $\gamma/\Gamma > 2$ are even harder to achieve since this would imply that the coherence time of the interband transition is significantly longer than that of the intraband one. This phenomenonology also implies that the current originating from $\eta_1 + \eta_2$ is directly related to the departure from adiabaticity, and in fact quantifies the degree to which the adiabatic approximation with the successive limits $\tau_1 \rightarrow \infty$ and then $\tau_0 \rightarrow \infty$ fails to hold.

For concreteness, we now use a relaxation time approximation for the temperature dependence of the relaxation rates, i.e. $\gamma = c_{00}(T) + c_{01}T^\alpha$, and $\Gamma = c_{10}(T) + c_{11}T^\alpha$. \hspace{1cm}
conductivity is eventually intercepted by localization effects due to disorder and phonon scattering is insensitive to the occupation numbers in valence and conduction bands, enforcing $\frac{\tau}{\gamma c} \ll 1$ to the occupied DOS in the densely populated valence band. However, for a strongly correlated system with low temperature dependence of both processes be essentially characterized by the same power of $T$, up to a numerical coefficient. In the usual fashion, the factors $c_{00}(0)$ describe the residual scattering induced by static disorder in the system. At low temperatures, the ratio approaches $\frac{\gamma}{\Gamma} = \frac{c_{10}(T)}{c_{00}(T)} \sim 1$, as the elastic mean free path due to disorder and phonon scattering is insensitive to the occupation numbers in valence and conduction band. However, for a strongly correlated system with low-disorder this will cross over into $\frac{\gamma}{\Gamma} = \frac{c_{11}}{c_{01}} \sim 2$ since the inelastic electron-electron interaction is proportional to the occupied DOS in the densely populated valence band, enforcing $\frac{\tau}{\gamma c} \rightarrow 0$. In both cases, the sub-gap conductivity is eventually intercepted by localization effects as a function of decreasing frequency. However, the onset of such activated behavior is markedly dissimilar. In the interacting case, decreasing the temperature will decrease both $\gamma$ and $\Gamma$ so that $\gamma/\Gamma = 2$, therefore the current decreases for any frequency below the gap (cf. Fig 2). In contradistinction, in the non-interacting case a decrease in temperature will again decrease both $\gamma$ and $\Gamma$ but with $\gamma/\Gamma \sim 1$. Then, the conductivity at frequencies between the mobility edge and the gap will remain unchanged. As the conductivity is qualitatively different for $\gamma/\Gamma < 2$ compared to $\gamma/\Gamma = 2$, one can determine whether or not the mobility edge - if it is observable - appears for states which are well described in a single-particle picture or rather ones which follow from a strongly coupled many-body description. We point out that the initial rapid decay of the conductivity for $\gamma/\Gamma = 2$ below the gap can make it difficult to detect a mobility edge for some compounds. Additionally, there are some restrictions on the suitable temperature range and the inelastic mean free path as the mobility edge is usually located a distance $V_0^2/\Lambda$ from the corresponding band edge, with $V_0$ the disorder strength and $\Lambda$ the bandwidth $[28–30]$.

Here, the first part is due to static disorder and electron-phonon interactions, while the $T^n$ piece is due to electron-electron interactions. Note that the power $n$ of the temperature dependence for both rates is taken to be the same, which is a good approximation as long as $\tau_1/\tau_0 > 1$. In this regime, we anticipate that the temperature dependence of both processes be essentially characterized by the same power of $T$, up to a numerical coefficient. In the usual fashion, the factors $c_{00}(0)$ describe the residual scattering induced by static disorder in the system. At low temperatures, the ratio approaches $\frac{\gamma}{\Gamma} = \frac{c_{10}(T)}{c_{00}(T)} \sim 1$, as the elastic mean free path due to disorder and phonon scattering is insensitive to the occupation numbers in valence and conduction band. However, for a strongly correlated system with low-disorder this will cross over into $\frac{\gamma}{\Gamma} = \frac{c_{11}}{c_{01}} \sim 2$ since the inelastic electron-electron interaction is proportional to the occupied DOS in the densely populated valence band, enforcing $\frac{\tau}{\gamma c} \rightarrow 0$. In both cases, the sub-gap conductivity is eventually intercepted by localization effects as a function of decreasing frequency. However, the onset of such activated behavior is markedly dissimilar. In the interacting case, decreasing the temperature will decrease both $\gamma$ and $\Gamma$ so that $\gamma/\Gamma = 2$, therefore the current decreases for any frequency below the gap (cf. Fig 2). In contradistinction, in the non-interacting case a decrease in temperature will again decrease both $\gamma$ and $\Gamma$ but with $\gamma/\Gamma \sim 1$. Then, the conductivity at frequencies between the mobility edge and the gap will remain unchanged. As the conductivity is qualitatively different for $\gamma/\Gamma < 2$ compared to $\gamma/\Gamma = 2$, one can determine whether or not the mobility edge - if it is observable - appears for states which are well described in a single-particle picture or rather ones which follow from a strongly coupled many-body description. We point out that the initial rapid decay of the conductivity for $\gamma/\Gamma = 2$ below the gap can make it difficult to detect a mobility edge for some compounds. Additionally, there are some restrictions on the suitable temperature range and the inelastic mean free path as the mobility edge is usually located a distance $V_0^2/\Lambda$ from the corresponding band edge, with $V_0$ the disorder strength and $\Lambda$ the bandwidth $[28–30]$.

Of course, when TRS is present it instead holds true that $\sigma \sim \Gamma$, resulting in a conductivity which carries no information about the ratio $\gamma/\Gamma$. In this latter case, one cannot separate the different sources of relaxation without exploring the microscopics of the electron motion. An effect which we neglected in the discussion so far is the broadening of the Fermi step for any non-zero temperature. Indeed, this is inconsequential as long as the gap $\Delta \gg k_B T$.

Conclusions.— In this letter, we investigated the second-order dc-conductivity in response to irradiation with light of frequency below the energy gap of a semiconductor or insulator lacking time-reversal symmetry. Our results show that the magnitude and frequency dependence of the conductivity is qualitatively different from the time-reversal symmetric case. Using a simple model for the quasiparticle relaxation, we proposed a way to probe the localization physics in experimentally realizable high-quality samples. Namely, we predict a large and temperature independent sub-gap dc-current for non-interacting quasiparticles and a $T$-dependent suppression of the bulk photovoltaic response for strong interactions. If a sub-gap mobility edge is observed in this latter case, it can be associated with localization originating from a many-body state.

The bulk photovoltaic effect has been studied only for few magnetic compounds $[31–34]$. Good material candidates to observe the proposed effect are transition metal dichalcogenide monolayers of MnP$x_3$, $X = Se, S$ $[35]$ and some hexagonal Mott insulators like RuCl$_3$ $[36, 37]$. Also, by measuring the second order conductivity above and below the Nel temperature, it is in principle possible to use the same sample to explore the response with and without TRS.
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