The bulk photovoltaic effect (BPVE) refers to current generation due to illumination by light in a homogeneous bulk material lacking inversion symmetry. Apart from the intensively studied shift current, the ballistic current constitutes an important contribution to the overall kinetic model of the BPVE. Ballistic current originates from asymmetric carrier generation due to scattering processes. In this Letter, using a perturbative approach, we derive a formula for ballistic current resulting from the intrinsic electron-phonon scattering in a form amenable to first-principles calculation. We then implement the theory and calculate the ballistic current of the prototypical BPVE material BaTiO$_3$ using quantum-mechanical density functional theory. The magnitude of the ballistic current is comparable to that of shift current, and the total spectrum (shift plus ballistic) agrees well with the experimentally measured photocurrents. Furthermore, we show that the ballistic current is sensitive to structural change, which could benefit future photovoltaic materials design.

**Keywords:** BPVE, shift current, ballistic current, first principle, electron-phonon coupling
that the magnitude of ballistic current can be significantly tuned.

Based on the Boltzmann transport equation, the phonon-mechanism ballistic current can be expressed as:

\[ j^{\alpha\beta}(\omega) = 2e\tau_0 \sum_{c\neq k} \Gamma^{\alpha\beta,asym}_{c\nu,k}(\omega)[v_{c\nu,k}^e - v_{c\nu,k}^e] \],

(1)

where \( \Gamma^{\alpha\beta,asym}_{c\nu,k}(\omega) \) is the asymmetric carrier generation rate for an electron-hole pair \((c,\nu)\) at \(k\), \(e\) is the electron charge, \(\tau_0\) is the momentum relaxation time, and \(v_{c\nu,k}^e\) is the electron (hole) velocity obtained from band derivatives. The leading factor of two is for spin degeneracy. The central quantity that needs to be evaluated is the asymmetric carrier generation rate, and it is derived below.

Adopting the velocity gauge \(E = -\frac{2A}{m}\) and taking the electron-photon interaction as \(\hat{H}_{e-photon} = \frac{e}{m}\hat{P}\cdot\hat{A}\), from linear response theory [28, 29], the average power delivered by monochromatic light of frequency \(\omega\) to the system during one period of oscillation is

\[ W = -2e\text{Im} \left[ \chi^{\alpha\beta}(\omega) \right] \left( \frac{e}{m\omega} \right)^2 E_\alpha(\omega)E_\beta(\omega), \]

(2)

where \( \chi^{\alpha\beta}(\omega) \) is the rank-two response function in the presence of \(E\) field with Greek letters denoting its components, \(e\) and \(m\) are the electron charge and mass, and \(E_\alpha(\omega)\) is the amplitude of the electric field, whose frequency dependence will be taken implicitly hereafter. Considering that each photon absorbed will be converted to an electron and hole, [23], the overall carrier generation rate \(\Gamma^{\alpha\beta}(\omega)\) can then be written as

\[ \Gamma^{\alpha\beta}(\omega) = \frac{W}{\hbar\omega} = \frac{2e}{\hbar} \text{Im} \left[ \chi^{\alpha\beta}(\omega) \right] \left( \frac{e}{m\omega} \right)^2 E_\alpha E_\beta. \]  

(3)

According to the Kubo formula, the response function is related to the retarded momentum-momentum correlation function:

\[ \chi^{\alpha\beta}(\omega) = \frac{1}{\hbar} G_R^{\alpha\beta}(\omega) \]

\[ = \frac{1}{\hbar} \int_{-\infty}^{+\infty} dt e^{-i\omega t} \Theta(t) \left\langle \left[ \hat{P}^\alpha(0), \hat{P}^\beta(t) \right] \right\rangle. \]

(4)

Here, the brackets \( \langle \cdot \rangle \) indicate an equilibrium average with respect to the total Hamiltonian that includes any extra interaction \(\hat{H}'\), which in our case is the electron-phonon interaction, and the momentum operators are in the Heisenberg picture. To evaluate \(\chi^{\alpha\beta}(\omega)\), we first calculate the imaginary-time (Matsubara) correlation function in its second quantization form with Bloch states as the basis:

\[ \chi^{\alpha\beta}_T(i\omega_n) = \frac{1}{\hbar \Lambda_{kk'\nu'\nu}} \sum_{c\nu,c'\nu'} \left\langle \nu k | \hat{P}^\alpha | \nu' k' \right\rangle \left\langle \nu' k' | \hat{P}^\beta | \nu k \right\rangle \times \int_0^{\Lambda/\hbar} d\tau e^{i\omega_n \tau} \left\langle \hat{T}_\tau c^\dagger_{c\nu}(\tau) \hat{c}_{c'\nu'}(\tau) \hat{c}_{c'\nu'}(0) \hat{c}_{c\nu}(0) \right\rangle, \]

(5)

where \( c(c') \) and \( \nu(\nu') \) are band indices for conduction and valence bands, respectively, \(k,k'\) are crystal momenta, and \(1/\hbar\Lambda T\) reflects the influence of temperature. The retarded and Matsubara correlation functions can be related through analytical continuation: \(\chi^{\alpha\beta}(\omega) = \chi_{\Lambda T}^{\alpha\beta}(i\omega_n \rightarrow \omega + i0^+)\), where \(+0^+\) is a infinitesimal positive number. In Eq. [5] two conditions hold: first, due to Pauli exclusion, transitions are only allowed from occupied valence bands to unoccupied conduction bands; also the population of electrons in a semiconductor is not significantly influenced by temperature, which indicates that the temperature for electrons can be taken as 0 K so that \(1/\hbar\Lambda T \rightarrow \infty\). Second, because of the negligible momentum carried by photons, only vertical transitions are allowed. From Eq. [3] it can be seen that the carrier generation rate \(\Gamma^{\alpha\beta}(\omega)\) can be decomposed into components from various \(k\) points and electron-hole pairs: \(\Gamma^{\alpha\beta}(\omega) = \sum_{c\nu,k} \Gamma^{\alpha\beta}_{c\nu,k}(\omega)\), and we only consider the asymmetric scatterings \(\Gamma^{\alpha\beta}_{c\nu,k}(\omega) \neq \Gamma^{\alpha\beta}_{c\nu,-k}(\omega)\) as the contribution to net current. Without any other interaction, Eq. [5] corresponds to Fermi’s golden rule, and this is a symmetric excitation which does not generate any current.

Therefore, we calculate the carrier generation rate in the presence of electron-phonon coupling. By introducing the Frölich e-ph Hamiltonian as [28, 30, 51]

\[ \hat{H}_{e-photon} = \sum_{\mu\nu\nu'} \sum_{kk'} g^{\mu\nu\nu'}_{kk'} c^\dagger_{\nu'} \hat{c}_{\nu} \hat{\Phi}^\mu_{k-k'} \]

(6)

where \(\hat{\Phi}^\mu_q = \hat{a}^\mu_q + \hat{a}^\dagger_{-q}\) is the phonon field operator, \(\hat{a}^\mu_q (\hat{a}^\dagger_{-q})\) are the phonon annihilation(creation) operators, and \(g^{\mu\nu\nu'}_{kk'}\) is the electron-phonon coupling matrix, we perform a perturbative expansion using a Feynman diagrammatic approach. The lowest-order non-zero contribution is second-order, illustrated as three different diagrams in Fig. [1]. As shown in the Supplementary Material, the processes of Fig. [1b] and [1c] are symmetric scattering, and only Fig. [1a] contributes to asymmetric scattering. By applying Feynman rules on Fig. [1a] and performing
analytical continuation, we can find the second-order correction to the carrier generation rate $\Delta \Gamma_{\text{carrier, } k}^{\alpha \beta} (\omega)$. Finally, we use relations that are satisfied for materials with time-

\[
\Gamma_{\text{carrier, } k}^{\alpha \beta, \text{sym}} (\omega) = \frac{1}{2} \left( \Delta \Gamma_{\text{carrier, } k}^{\alpha \beta} (\omega) - \Delta \Gamma_{\text{carrier, } -k}^{\alpha \beta} (\omega) \right) = \frac{2}{h} \left( \frac{\pi e^2}{m} \right)^2 E_\alpha E_\beta \sum_{c'v'k'} \text{Im} \left[ \left\langle v | k \right| \hat{P}^\alpha | c', -k \right\rangle \left\langle c' | \hat{P}^\beta | v', k \right\rangle g_{\mu \nu}^{\alpha \beta} g_{\mu \nu}^{c'c} \right] \\
\times \left\{ (N_\mathbf{q} + 1) \left[ \delta(E_{ck} - E_{ck} - h\omega) \delta(E_{c'k'} - E_{v'k'} - h\omega) \right] \left( \frac{1}{E_{c'k'} - E_{ck} + h\omega_\mathbf{q}} + \frac{1}{E_{ck} - E_{v'k'} + h\omega_\mathbf{q}} \right) + \delta(E_{ck} - E_{ck} - h\omega) \frac{1}{E_{c'k'} - E_{ck} + h\omega_\mathbf{q}} \left[ \delta(E_{c'k'} - E_{ck} + h\omega_\mathbf{q}) + \delta(E_{c'k'} - E_{ck} + h\omega_\mathbf{q}) \right] + N_\mathbf{q} [\omega_\mathbf{q} \leftrightarrow -\omega_\mathbf{q}] \right\} 
\]  

(8)

where $\mathbf{q} = \mathbf{k} - \mathbf{k}'$ is the phonon momentum, $N_\mathbf{q}$ is the phonon population, and $[\omega_\mathbf{q} \leftrightarrow -\omega_\mathbf{q}]$ denotes the term in brackets in Eq. 8 with instances of $\omega_\mathbf{q}$ negated. The delta functions in Eq. 8 reflect the selection rule for optical transition, and the electron-phonon coupling matrices together with principal parts are the modulation of the transition rate. The initial asymmetric carrier distribution quickly thermalizes, so the carriers contribute to the current only for times on the order of the momentum relaxation time of the carriers, which is usually on the femtosecond time scale. We approximate $\tau_0$ to be 4 fs in this work, which is justified by an estimation from first-principles calculations (See SI). Together, Eq. 1 and Eq. 8 provide a method to compute the ballistic current density from quantities that are readily available from first-principles calculations.

We perform density functional theory (DFT) and density functional perturbation theory (DFPT) calculations using the QUANTUM ESPRESSO package [32, 33]. Generalized gradient approximation exchange correlation functional and norm-conserving pseudopotentials produced by the OPIUM package are used [34, 35]. The convergence threshold for self-consistent calculations was $10^{-8}$ Ry/cell, and for DFPT calculations it was $10^{-16}$ Ry/cell. Velocity and electron-phonon coupling matrices are calculated by Wannier interpolation using the EPW package [37, 38]. All quantities are sampled on an $8 \times 8 \times 8$ unshifted Monkhorst-Pack grid [39], and the principal part integration is dealt with using a generalized Newton-Cotes method (See SI).

BaTiO$_3$, as a prototypical ferroelectric and bulk photovoltaic material, is an ideal candidate for benchmarking the ballistic current; the BPVE current spectrum has been measured for BaTiO$_3$ [27], and the shift current has also been predicted by first-principles calculations [7]. We use the experimental lattice parameters of tetragonal BaTiO$_3$ with Ti-displacement along (001) to represent the spatially-averaged structure, and the atomic positions are relaxed before the phonon calculations. The theoretical ballistic current is shown in Fig. 2(a). We find that the ballistic current has a more jagged response profile, which is indicated by [20] as a signature of the ballistic current. For the considered range of light frequency, the largest calculated response occurs at 2.1–2.5 eV above the band gap, similar to the shift current (Fig. 2(c)). Even though the lineshape of the ballistic current is more complicated, we note that the turn-on frequency of $\sigma_{zzZ}$ is larger than that of $\sigma_{xxZ}$ for both ballistic and shift current. In addition, the amplitudes of the ballistic and shift current and similar in magnitude, and thus we find that both shift current and ballistic current will contribute significantly to the experimentally measured current.

To compare with experiment, we calculate the real photocurrent based on the Glass coefficient [18, 40], by further computing the absorption coefficient with the quasi-particle correction. As pointed out by our previous work [18], the quasi-particle correction will significantly influence the absorption profile, but it will mainly blue-shift the response tensor within the frequency range of interest. We apply the same technique by calculating the absorption coefficient using quasi-particle energies with exciton correction, while still keeping the velocity matrix and electron-phonon coupling matrix at the GGA.
level. In this way, accuracy is improved while the computational cost is kept low. In Fig. 3, the $xxZ$ ballistic current partially fills the gap between the shift current and the experimental spectra, whereas for the $zzZ$ component whose shift current has already aligned well with the experiments, the ballistic current barely influence the theoretical BPVE spectrum. This confirms that the ballistic current from the electron-phonon scattering can contribute significantly to the BPVE. However, we want to point out that in order to get a full understanding of the ballistic current and the BPVE, other scattering mechanisms such as defect scattering and electron-hole Coulomb scattering should also be taken into account.

As revealed by previous study, the shift current response can be strongly enhanced by modest changes to crystal structure or composition [41, 42]. Here, we extend this idea and explore the relation between the ballistic current and structure. We find that certain structures can greatly enhance the current response. To illustrate this point, we lift all constraints of BaTiO$_3$ and perform a full structural relaxation, so that the low-temperature rhombohedral phase is obtained. For this low-temperature structure, the corresponding ballistic current photovoltaic tensor is shown in Fig. 2(b). Its lineshape is dramatically different from that of the tetragonal phase (Fig. 2a), and the overall magnitude is much larger. Through a visual inspection of the two structures (the insets of Fig. 2(a) and (b)), we find a larger off-center displacement along the (111) direction in the rhombohedral structure and a smaller distortion along the (100) direction in the tetragonal phase. This could indicate a relation between the magnitudes of the current response and the structure distortion. Specifically, it could be that a larger extent of symmetry breaking will enhance the
asymmetry of the momentum distribution, and the off-center displacement suggests that different parts of the Brillouin zone will not contribute to the ballistic current uniformly. A more quantitative investigation into the relationship between structure and the ballistic current will be the topic of our future study. For practical applications, however, this contrast between the ballistic current responses of rhombohedral and tetragonal BaTiO$_3$ is very illuminating since it shows that a large part of the solar spectrum can be harvested by engineering the distortion via doping or external strain.

In conclusion, based on the Kubo formula, we derived an expression for the phonon-assisted ballistic current, and we implement it into first-principles calculation. Taking BaTiO$_3$ as an example, we demonstrated that the ballistic current can be an important mechanism for the BPVE. We showed that, similar to the shift current, the ballistic current is also very sensitive to structures; this reflects a promising possibility of material engineering to further harvest BPVE.

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Ballistic Current From First Principle Calculations: Supplementary Information

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DETAILED DERIVATION OF EQ. 7

By applying Feynman rule[1] in the momentum-frequency space on FIG.1(a) in the main text, we can get:

$$
\Delta \chi^{(2)\alpha\beta}(\omega) = \frac{1}{\hbar} \sum_{kk'cc'vv'} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k'
$$

$$
\times - \frac{1}{\hbar^2} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \frac{d\omega''}{2\pi} G_c(\omega', k) G_v(\omega', k) G_c(\omega'', k') G_v(\omega'', k') D_\mu(\omega' - \omega'', k - k') \quad (S1)
$$

Here, $G_c(\omega, k)$ and $G_v(\omega, k)$ are the bare electron and hole Matsubara’s function, and $D_\mu(\omega' - \omega'', k - k')$ is the free phonon Matsubara’s function. Notice that in a general Matsubara’s function formalism, the internal frequencies are usually summed over discrete frequency points instead of continuous integrals. However, since we are only considering the 0 K condition for electrons and the temperature influence on the phonon can be reflected by the population factor, we replace the summations with integrals. The next step is to do the analytical continuation: $i\omega = \omega + i0^+$, which is equivalent by replacing the Matsubara’s functions with the real-time Green’s functions. We note that strictly speaking, the replacement at this step would not necessarily yield the same result as the analytical continuation after the frequency summation of the Matsubara’s functions, but it can be shown to be valid for $\beta \to \infty$, which corresponds to 0 K situation. Then, we can get the second-order correction to the retarded correlation function and thus to the susceptibility:

$$
\Delta \chi^{(2)\alpha\beta}(\omega) = \frac{1}{\hbar} \sum_{kk'cc'vv'} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k'
$$

$$
\times - \frac{1}{\hbar^2} \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \frac{d\omega'}{2\pi} \frac{d\omega''}{2\pi} G_c(\omega', k) G_v(\omega', k) G_c(\omega'', k') G_v(\omega'', k') D_\mu(\omega' - \omega'', k - k') \quad (S2)
$$

where[2]

$$
G_c(\omega, k) = \frac{1}{\omega - E_{ck}/\hbar + i0^+},
$$

$$
G_v(\omega, k) = \frac{1}{\omega - E_{vk}/\hbar - i0^+},
$$

$$
D_\mu(\omega, k) = (N_q + 1) \left( \frac{1}{\omega - \omega_q + i0^+} - \frac{1}{\omega + \omega_q - i0^+} \right) + N_q \left( \frac{1}{\omega + \omega_q + i0^+} - \frac{1}{\omega - \omega_q - i0^+} \right) \quad (S3)
$$

Combining Eq. S2 and Eq. S3, we proceed by applying the residue theorem sequentially on $\omega'$ and $\omega''$, which will yield:

$$
\Delta \chi^{(2)\alpha\beta}(\omega) = - \sum_{kk'cc'vv'} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k'
$$

$$
\times \{ (N_q + 1) \frac{1}{E_{ck} - E_{ck} - \hbar - i0^+} \frac{1}{E_{ck} - E_{ck} + \hbar - i0^+} \frac{1}{E_{ck} + E_{ck} - \hbar - i0^+} \frac{1}{E_{ck} + E_{ck} + \hbar - i0^+} + N_q [\omega_q \leftrightarrow -\omega_q] \}
$$

$$
= - \sum_{kk'cc'vv'} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k' I(c, c', v, v', k, k', \mu) \quad (S4)
$$

Thus, the second-order carrier generation rate for an electron-hole pair can be written as:

$$
\Delta \Gamma_{cv,h}(\omega) = - \frac{2}{\hbar} Im[\Delta \chi^{(2)\alpha\beta}(\omega)] \left[ \frac{e}{m_\omega} E(\omega) \right]^2 = \frac{2}{\hbar} \left( \frac{eE(\omega)}{m_\omega} \right)^2 Im \left[ \sum_{k'v'c'\mu} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k' I(c, c', v, v', k, k', \mu) \right] \quad (S5)
$$

Considering Eq. 6 and the fact that $I(c, c', v, v', -k, -k', \mu) = I(c, c', v, v', k, k', \mu)$ as the time-reversal symmetry makes $E(n, k) = E(n, k)$ and $\omega_q = \omega_q$, we are able to get the asymmetric carrier generation:

$$
\Gamma_{cv,k}^{\alpha\beta, asym}(\omega) = \frac{1}{2} (\Delta \Gamma_{cv,k}^{\alpha\beta}(\omega) - \Delta \Gamma_{cv,-k}^{\alpha\beta}(\omega)) = \frac{2}{\hbar} \left( \frac{eE(\omega)}{m_\omega} \right)^2 Im \left[ \sum_{k'v'c'\mu} \langle vk|P^\alpha|ck \rangle \langle c'k'|P^\beta|v'k' \rangle g^{cc'}_{\mu} g^{vv'}_{\mu} k k' k k' \mu \mu k k' I(c, c', v, v', k, k', \mu) \right] \quad (S6)
$$
We can still move forward a little bit by substituting
\[ \frac{1}{\omega + i0^+} = \frac{1}{\omega} \mp i\pi\delta(\omega) \] (S7)
into \( I(c,c',v,v',k,k',\mu) \). After collecting the terms that are real and satisfy the selection rule for optical transition, we can finally get Eq. 7 in the main text. If, however, the system possesses inversion symmetry, then the additional equalities will hold up to a phase:
\[ \langle v, -k | P^\alpha | c, -k \rangle = -\langle v, k | P^\alpha | c, k \rangle \]
\[ g_\mu^{\alpha\beta}_{c,c'} = g_\mu^{\alpha\beta'}_{c',c} \] (S8)
and these equalities will make the asymmetric generation rate vanish.

PROOF THAT OTHER DIAGRAMS DO NOT CONTRIBUTE TO THE ASYMMETRIC SCATTERING

Since the algebraic structure of FIG.1(b) and FIG.1(c) will be the same and they can be obtained from each by a relabeling \( c \leftrightarrow v \), it suffices to only prove that FIG.1(b) has no contribution to asymmetric carrier generation. Following the same procedure for FIG.1(a), we can get the second-order correction to carrier generation from FIG.1(b) as:
\[ \Gamma^{\alpha\beta}_{cc,k}(\omega) = N\mbox{Im} \left[ \sum_{k'c'v'\mu} \langle vk | P^\alpha | ck \rangle \langle ck | P^\beta | vk \rangle |g_\mu^{c'c}_{kk'}|^2 I^{(b)}(c,c',v,k,k',\mu) \right] \] (S9)
Here, the additional superscript \( (b) \) indicates that it represents the contribution from FIG.1(b), and \( N \) is the prefactor, which is a real constant. \( I^{(b)}(c,c',v,k,k',\mu) \) is the result after the frequency integrals of the Green’s function. Similar to the \( I(c,c',v,v',k,k',\mu) \) in Eq. S4, it is a function of the electronic energies and the phonon energies, which will be unchanged under the change from \( k \) to \(-k\). To proceed, we notice that \( \Gamma^{\alpha\beta}_{cc,k}(\omega) \) and \( \Gamma^{\alpha\beta}_{cc,k}(\omega) \) will be inseparable in experiments, and therefore they should always be considered simultaneously:
\[
\left[ \Gamma^{\alpha\beta}_{cc,k}(\omega) + \Gamma^{\alpha\beta}_{cc,k}(\omega) \right] - \left[ \Gamma^{\alpha\beta}_{cc,-k}(\omega) + \Gamma^{\alpha\beta}_{cc,-k}(\omega) \right] \\
= N\mbox{Im} \left\{ \sum_{k'c'v'\mu} |g_\mu^{c'c}_{kk'}|^2 I^{(b)}(c,c',v,k,k',\mu) \times \left[ \langle vk | P^\alpha | ck \rangle \langle ck | P^\beta | vk \rangle + \langle vk | P^\beta | ck \rangle \langle ck | P^\alpha | vk \rangle - \langle vk | P^\alpha | ck \rangle^* \langle ck | P^\beta | vk \rangle^* - \langle vk | P^\beta | ck \rangle^* \langle ck | P^\alpha | vk \rangle^* \right] \right\}
\]
\[ = 0 \] (S10)
Thus, we have proved that FIG.1(b) has no contribution to the asymmetric carrier generation, and so does FIG.1(c).

ESTIMATION OF THE MOMENTUM RELAXATION TIME \( \tau_0 \)

We use the as-generated electron-phonon coupling matrices for tetragonal phase BaTiO\(_3\) to estimate the momentum relaxation time according to the formula in [3]. As can be seen in Fig. S1, the statics of all the momentum relaxation times at different \( (c,k) \) are very narrowly distributed. We point out that other scattering mechanism can also contribute to the momentum relaxation, which could possibly make the distribution even narrower. Therefore, it is a good approximation to take \( \tau_0 \) as a constant throughout. As we are going to compare the current density at the band edge with experiments, we chose the average momentum relaxation times of band edge states (states around Gamma point) as our \( \tau_0 \) in the ballistic current calculation, which yields \( 4.071 \times 10^{-15} \) s. Thus, we approximate \( \tau_0 = 4 \) fs.

GENERALIZED NEWTON-COTES METHOD

Numerically, for principal part (PP) integrals like Eq. 8, the common practice is to add a small pure imaginary number in the denominator to ‘smear’ it, but it usually requires a very dense k-grid to get a convergent result.
FIG. S1. Distribution of the momentum relaxation time of different (c, k)

However, since our final expression Eq. 1 has a double sum over k, it is computationally very expensive both to get the required ingredients and to perform the summation itself for a denser k-grid, which will make such calculation prohibitive. To circumvent this problem, we devised a new numerical PP integral scheme inspired by [4]. For the most general case, we are dealing with a three-dimensional integral in the form of:

$$I = \mathcal{P} \int_{V} d^{3}k \frac{f(\vec{k})}{g(\vec{k})} - p$$

(S11)

where p is a parameter and V represents the spatial region where the integration is performed. Our scheme is to split the overall region into several regions that are small enough:

$$I = \sum_{\Delta V} I(\Delta V) = \sum_{\Delta V} \mathcal{P} \int_{\Delta V} d^{3}k \frac{f(\vec{k})}{g(\vec{k})} - p.$$

(S12)

Since each integration region is small, we are allowed to perform the multi-variate Taylor expansion on $f(\vec{k})$ and $g(\vec{k})$ without loss too much of the accuracy:

$$I(\Delta V) \approx \mathcal{P} \int_{\Delta V} d^{3}k \frac{f(\vec{k}_{0}) + \frac{\partial f}{\partial k_x} k_{x0} (k_x - k_x0) + \frac{\partial f}{\partial k_y} k_{y0} (k_y - k_y0) + \frac{\partial f}{\partial k_z} k_{z0} (k_z - k_z0)}{g(\vec{k}_{0}) + \frac{\partial g}{\partial k_x} k_{x0} (k_x - k_x0) + \frac{\partial g}{\partial k_y} k_{y0} (k_y - k_y0) + \frac{\partial g}{\partial k_z} k_{z0} (k_z - k_z0) - p}$$

(S13)

In this small region $\Delta V$, $k_x0$, $k_y0$, $k_z0$, $f(\vec{k}_{0})$, $g(\vec{k}_{0})$, and the partial derivatives calculated from two-points formula can be taken as constants, so we are left with two types simple integrals, which has analytical forms for a cuboid region:

$$\mathcal{P} \int_{x_{1}}^{x_{2}} \int_{y_{1}}^{y_{2}} \int_{z_{1}}^{z_{2}} dxdydz \frac{1}{Ax + By + Cz - p'}$$

(S14)

$$\mathcal{P} \int_{x_{1}}^{x_{2}} \int_{y_{1}}^{y_{2}} \int_{z_{1}}^{z_{2}} dxdydz \frac{x}{Ax + By + Cz - p'}$$

(S15)

where A, B, C and $p'$ are constants. By doing these integrations analytically, we can get the integral for the volume element $\Delta V$, of which the summation would yield the the overall value of the integral. We can see that this method is fact a generalized Newton-Cotes(GNC) method and the analytical integration over the poles will be expected to eliminate the numerical instability.
FIG. S2. The comparison between the generalize Newton-Cotes method and the smearing method for integrals in Eq. S16. \( \gamma \) is the value of the small imaginary number in the smearing method. (a) \( p = 0.6 \). (b) \( p = 0.4 \). The smearing method requires a very dense grid to give well-converged results, but the GNC method can yield converged results even for a very small number of grid points. In addition, the GNC method is not sensitive to the position of the poles as it can give fast convergence for both \( p = 0.6 \) and \( p = 0.4 \). For the smearing method, however, even though when \( p = 0.6 \), various smearing values can yield convergent result given denser grids as shown in (a), only \( \gamma = 0.01 \) will give rise to convergent result when \( p = 0.4 \), which can be seen in (b).

We test the GNC method against the smearing method by doing both on a simple integral with two variables which can be done analytically:

\[
f(p) = \mathcal{P} \int_{-0.5}^{0.5} \int_{-0.5}^{0.5} dx dy \frac{1}{x^2 + y^2 - p}
\]

and the results are shown in FIG.S2. As expected, the traditional smearing method requires a very dense grid to give well-converged results, but the GNC method can yield converged results even for a fairly small number of grid points. Another advantage of the GNC method over the smearing method is its insensitivity to the position of the poles. In FIG.S2(a) where \( p = 0.6 \), various smearing values can yield convergent result given denser grids, whereas when \( p = 0.4 \) as shown in FIG.S2(b), only \( \gamma = 0.01 \) will give rise to convergent result. Notably, however, the GNC method can give fast convergence for small number of grid points in both cases. Therefore, in our calculations, we choose the GNC method to evaluate Eq. 7 and Eq. 8.

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