THz radiation of jerk photocurrent

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We compute the jerk current tensor of prototypical semiconductors GaAs, Si, and novel ferroelectric single-layer GeS, GeSe, SnS, and SnSe. We find peak jerk current tensor values of the order of $10^{14}$ mA/V$^3$s$^2$ in GaAs and Si in the visible energy spectrum and an order of magnitude larger in single-layer GeS, GeSe, SnS, and SnSe. We attribute the large magnitude of the response in the latter to the restrictions imposed by dimensionality and symmetry. More important, the detailed knowledge of this tensor and its large value in single-layer GeS, GeSe, SnS, and SnSe make it possible to predict the magnitude and angle of rotation of polarization of intense THz pulses generated in photoconductive switches and point to new functionalities of these devices not explored before.

**Introduction.** Understanding and controlling light-matter interactions is at the forefront of scientific discovery and technological applications. This is specially true for nonlinear optical effects. The second harmonic generation,[1] for example, is routinely used as a frequency multiplier, as a probe of material’s crystal symmetry (or lack of), as enabler of entangled photons in quantum protocols, etc. Recently, the bulk photovoltaic effect (BPVE)[2–38], i.e., the generation of dc current in illuminated insulators lacking inversion symmetry, has attracted renewed attention for its possible application in optoelectronics, in particularly using topological insulators,[29] novel two-dimensional (2D) ferroelectric materials [9–15], and Weyl semimetals [30–32]. Traditionally, the BPVE refers to a second order effect in the optical field but it was recently extended to higher orders giving rise to novel BPVEs effects and to an explicit expression for the photoconductivity, i.e., intensity dependent conductivity, in terms of Bloch wavefunction parameters of the crystal.[37, 38]

The photocconductivity has long been studied in the context of generation of intense terahertz (THz) pulses in photoconductive dipole switches (Hertzian dipoles).[39, 40] A typical photoconductive (dipole) switch consists of two metal electrodes on a semiconductor substrate (usually low temperature GaAs) separated by a distance (100 μm to cms), see Fig. 1. A potential difference between these fields establishes a static electric field $E_0$. An optical pulse of femtosecond (fs) duration is incident in the gap between the electrodes. The photocurrent generated radiates an electric field in the THz frequency range given by [41]

$$E_{thz} \propto \frac{dJ}{dt},$$

(1)

far from the source. Usually, the pulse has a gaussian shape with a central frequency just above the energy band gap. The carriers are assumed to form a free gas of electrons with the total number of carriers available for conduction proportional to the intensity of the pulse envelop. The dynamics of the driven gas has been stud-

**Jerk current.** Consider an optical $\mathbf{E}$ and a static field $\mathbf{E}_0$. An expansion of the photocconductivity in powers of these fields gives three contributions [37, 38] which schematically we can write as

$$J_{dc, ph}^{(3)} = \epsilon_3 \mathbf{E}^2 \mathbf{E}_0 + \eta_3 (\mathbf{E} \times \mathbf{E}^*) \mathbf{E}_0 + \sigma_3 \mathbf{E}^2 \mathbf{E}_0.$$

(2)

The first term is the jerk current and is expected to be the largest. The second and third terms are higher-order versions of the injection and shift currents of the BPVE. For a monochromatic and spatially homogeneous field $\mathbf{E}^\omega \equiv E_0^\omega e^{-i\omega t} + c.c.$ the jerk current obeys the phenomenological equation [37]

$$\frac{d^2}{dt^2} J_{jerk}^{(3)} = 6\epsilon_3 \omega \delta \delta_0 (0, \omega, -\omega, 0) E_0^F E_0^c E_0 E_0 \left( \frac{1}{T^2} J_{jerk}^{(3)} \right).$$

(3)

FIG. 1. The photo conductive switch shown in profile. An incident pulse with central frequency above the energy band gap (red) produces a single-cycle pulse in the THz frequency regime (green).
The first term represents a process by which carriers pumped into conduction states accelerate uniformly under a static electric field $dv_n^a/dt = \omega_{nm}E_0^d$. The static field picks a direction and breaks the symmetry between $\pm k$ states for both for linear and circular polarization. It is well known that the rate of carrier injection can be calculated from Fermi’s golden rule and for elliptical polarization we have $df_n(-k)/dt \neq df_n(k)/dt$ leading to injection current.[6, 38]

The second term represents a process by which the carrier injection rate itself changes in time. Since the wave vector of a wave packet evolves as $eE_0/t$ in the presence of the static field, the time-reversed states $\pm k$ are ‘Doppler’ shifted to $\pm k + eE_0/t$ in the frame of reference where a wave packet is at rest. In this frame the field is zero $E_0 = 0$ and the degeneracy between $\pm k$ states is lifted in the sense that

$$\frac{d^2}{dt^2}f_n(-k) \neq \frac{d^2}{dt^2}f_n(k),$$

(7)

giving rise to a current, see supplemental information (SI). Note again, that this process is very different from the injection current where the carrier injection rates at $\pm k$ are asymmetric only for circular or elliptic polarization. The jerk current, on the other hand, can be finite for both polarizations.

**Jerk current tensor in GaAs and monolayer GeS.** GaAs point group, 43m, allows three independent components of $\iota_{3}^{abcd}$, xyyx, xxyy, and xxxx shown in Fig. 2 as a function of incoming photon energy (frequency). The numerical details of the density functional theory (DFT) calculation are presented in the SI. The jerk spectrum vanishes for photon energies less than the energy band gap of 1.4 eV. The spectrum peaks at 3 eV, which lies in the visible spectrum, and at 4.7 eV, both peaks are of the order of $6 \times 10^{14}$ mA/V s$^2$ and $12 \times 10^{14}$ mA/Vs$^2$. Note that current transverse to the static field is possible and is controlled by the xxyy component but it is about an order of magnitude smaller with respect to the longitudinal components. As shown in the SI, the isolated peaks in $\iota_3$ can be explained, in part, by the high joint density of states (JDOS) at various points in the Brillouin zone (BZ). The response tensor for Si has a similar spectrum and is also presented in the SI.

It is interesting to compare the spectrum of $\iota_3$ of GaAs, which breaks inversion symmetry but is not ferroelectric, with monolayer GeS, which is predicted to have large in-plane spontaneous polarization, [13, 47] and with Si which does not break inversion symmetry and hence is not ferroelectric. Let us chose the slab to define the in-plane spontaneous polarization we have $E_0 = 0$.

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not considered here. The 2D values are reported as bulk equivalent, i.e., per unit volume to be able to compare easily with other materials. The spectrum of monolayer GeSe, SnSe, and SnSe is qualitatively similar and are presented in the SI.

The spectrum of $\ell_3$ in monolayer GeS is zero for frequencies smaller than the energy gap 1.9 eV, as expected. In contrast to GaAs and Si, the response is flatter over range of photon energies 2.7-5.5 eV which includes part of the visible spectrum. Importantly, the peak responses are an order or magnitude larger than in GaAs and Si, and can reach peak values of $70 \times 10^{14}$ mA/V^3s^2 in monolayer SnSe (see SI). The transverse components $y_yxx$ and $x_yxy$ are an order of magnitude smaller than the longitudinal components. Analysis of the JDOS, spin-orbit coupling (SOC) and polarization suggest that the reduced dimensionality, the symmetries imposed by the in-plane polarization and JDOS are responsible for the larger response. A similar conclusion was reached in the case of injection current in these materials.[15]

**THz radiation of jerk current.** We now demonstrate that the plane of polarization of the emitted THz pulse is rotated with respect to the direction of the bias field. We consider a pulse envelop width $w$ much larger than the period the central frequency $T$. In such case, many cycles occur at approximately constant amplitude. We also assume the momentum relaxation time $\tau$ is small compared with $w$ but larger compared to $T$ as

$$w \gg \tau \gg T. \quad (8)$$

Under such conditions, it is easy to see that the magnitude of the instantaneous (time varying) photocurrent is proportional to the (time varying) intensity of the envelope times the jerk current tensor evaluated at the frequency of the central carrier, Eq. 4. In practice, these assumptions are not stringent since for typical clean semiconductors with an energy band gap in the visible $T \sim 2$ fs, $\tau \sim 50$ fs and $w \sim 200$ fs, see for example Refs.[48–50]. In addition, we assume the injected carriers do not saturate the semiconductor. This is usually the case for moderate (but experimentally relevant) bias fields and optical powers.[42] The advantages of working in this regime are: (i) the frequency dependance of $\ell_3$ provides useful information about the magnitude of the photocurrent response and hence of the THz field amplitude. For example, our calculation of $\ell_3$ above suggests that photon energies of 3 eV produce twice as much (jerk) photocurrent in GaAs than at the band edge at 1.4 eV. Previous experiments almost always use photon energies just above the band edge.[45] More important, monolayer GeS produces an order of magnitude larger (jerk) photocurrent than GaAs and Si. (ii) Lattice anisotropy could give rise to THz radiation not along the bias field, an effect not captured in Drude-like models where the emitted THz field is parallel to $E_0$.

As a first example, consider an incident optical field $E$ perpendicular to the ab-plane of GaAs which is assumed to coincide with the lab’s xy-plane (see Fig. 1) and a static in-plane field $E_0$

$$E = \hat{x}E_x^0 e^{-i\omega t} + \hat{y}E_y^0 e^{-i\omega t} + c.c., \quad (9)$$
$$E_0 = \hat{x}E_x^0 + \hat{y}E_y^0, \quad (10)$$

where $E_x^0 = |E_x^0|e^{-i\phi_x}$, $E_y^0 = |E_y^0|e^{-i\phi_y}$. The angle of the current will define the angle of the THz pulse polarization. Let us define $\theta_{thz}$ with respect to the $x$-axis as

$$\tan \theta_{thz} = \frac{J_y^{\text{jerk}}}{J_x^{\text{jerk}}}. \quad (11)$$

Explicit calculation gives for GaAs

$$\theta_{thz} = \theta_{E_0}, \quad \text{(circ. pol.)}, \quad (12)$$
$$\tan \theta_{thz} = \frac{2\ell_{xy}}{\ell_{xx} + \ell_{yy}} \quad (45 \text{ deg pol. } E_0 \parallel \hat{x}) \quad (13)$$

where $\theta_{E_0}$ is the angle of the bias field. The first result agrees with the predictions of isotropic models.[45] The

![Graph showing THz radiation of jerk current](image-url)
the angle is independent of the strength of the static and THz pulses in monolayer GeS. The polar axis defines which could be generated, vanishes for the geometry we consider. which THz output power is largest with lowest bias voltage, subsequent thermal load and saturation effects. In addition, our calculations predict large rotation of the polarization plane of the emitted THz pulse, making it possible to produce on-demand THz pulses with a given polarization by either changing the bias voltage, the frequency, or both. In addition, jerk current contributes to linear polarization[13] with an angle \( \tan \theta_{thz, shift} = \frac{2\sigma_2^{thz}}{(\sigma_2^{thz} + \sigma_2^{circ})} \) for typical static fields the jerk current is expected to be much larger. An estimate of the relative magnitudes of these currents was given in Ref.[38]

**Conclusions.-** In this paper we use DFT to compute the jerk current response tensor \( \epsilon_{abc}^{thz} \) of prototypical semiconductors GaAs and Si and novel ferroelectric single-layer GeS, GeSe, SnS and SnSe as a function of photon energy. The jerk current spectrum of GaAs and Si is composed of isolated peaks, largely explained by high JDOS, and which can reach values up to \( 6 \times 10^{14} \text{mA/V}^3 \text{s}^2 \) in the visible spectrum. The spectrum of monolayer GeS, GeSe, SnS and SnSe, on the other hand, is flatter over a large range photon energies (including the visible frequency regime) with peak values about an order of magnitude larger than those in GaAs and Si.

Jerk current processes are expected to be the largest contribution to photoconductivity and hence can be used to learn about the intensity and polarization of intense THz pulses emitted by photoconductive switches. Our calculation builds on isotropic models and incorporate anisotropic effects of the lattice. The detailed knowledge of \( \epsilon_{abc}^{thz} \) makes it possible to predict the frequency at which THz output power is largest with lowest bias voltage. This provides a solution to the main drawback of Hertzian dipole technology, namely, degradation of emitters due to high bias voltage, subsequent thermal load and saturation effects. In addition, our calculations predict large rotation of the polarization plane of the emitted THz pulse, making it possible to produce on-demand THz pulses with a given polarization by either changing the bias voltage, the frequency, or both. In addition, jerk current does not require breaking of inversion symmetry and hence a wide range materials can be used.

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**FIG. 4.** (a) Angle of polarization of emitted THz pulse in a GaAs Hertzian dipole. The optical field is polarized at 45 deg and the static field is \( \mathbf{E}_0 = \hat{x} E_{0x}^d \). Isotropic models predict a response parallel to \( \mathbf{E}_0 \) at all frequencies (dashed lines). Note that the angle of the emitted THz pulse does not depend on the magnitude of the static or optical fields nor on the details of the relaxation mechanism. (b) Same but for monolayer GeS. Two polarizations are indicated along with the isotropic model predictions (dashed lines). For circular polarization of light, the static field is at 45 deg. For linear polarization of light, \( \mathbf{E}_0 = \hat{x} E_{0x}^d \).

second result, obtained for a linearly polarized optical field at an angle of 45 deg and \( \mathbf{E}_0 = \hat{x} E_{0x}^d \), is different from \( \theta_{thz} = 0 \) predicted by isotropic models. Note that, the angle is independent of the strength of the static and optical fields and of the microscopic parameter \( \tau \) as long condition 8 is satisfied. As such, it is an intrinsic property of the Bloch wavefunction of the material. The amplitude of the emitted THz field is proportional to the intensity of pulse envelop and the bias field. Naturally, the power emitted is proportional to the square of the bias field. Specifically, the power emitted is \( \frac{1}{4} \left( \frac{\sigma_2^{thz}}{\sigma_2^{circ}} + 1 \right) |E_0|^2 \) for typical static fields the jerk current is expected to be much larger. An estimate of the relative magnitudes of these currents was given in Ref.[38]

(see Fig. 1). Contrary to GaAs, the angle of rotation for two representative cases is

\[
\tan \theta_{thz} = \frac{\nu_{xy}^{thz} + \nu_{yxy}^{thz}}{\nu_{xxy}^{thz} + \nu_{yy}^{thz}} \tan \theta_{E_0} \quad \text{(circ. pol.)} \quad (14)
\]

\[
\tan \theta_{thz} = \frac{2\nu_{xy}^{thz}}{\nu_{xxy}^{thz} + \nu_{yy}^{thz}} \quad \text{(45 deg pol. \( \mathbf{E}_0 \parallel \hat{x} \))} \quad (15)
\]

and is shown in Fig. 4(b). Again, the angles differ from the predictions of isotropic models. In fact, the polarization can turn almost a full 360 deg for frequencies near the band edge in the visible regime. Since monolayer GeS breaks inversion symmetry injection and shift current will be generated. The injection current contributes \( J_y^{inj} = 4i\tau \nu_{yxy}^{thz} |E_0|^2 \) to circular polarization.[15] In typical experimental setups the static field \( 10^5 \text{ V/m} \) will generate a jerk current much larger than the injection current. Shift current contributes to linear polarization[13] with an angle \( \tan \theta_{thz, shift} = \frac{2\sigma_2^{thz}}{(\sigma_2^{thz} + \sigma_2^{circ})} \), but for typical static fields the jerk current is expected to be much larger. An estimate of the relative magnitudes of these currents was given in Ref.[38]
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[54] TINIBA is a tool written in bash, perl, and fortran.
SUPPLEMENTAL INFORMATION: THZ RADIATION OF JERK PHOTOCURRENT

Classical contribution to jerk current

The jerk current admits a simple semiclassical interpretation. To see this take two time derivatives of

\[ J = \frac{e}{V} \sum_{nk} f_n v_n, \]  

and keep terms to third order in the electric field

\[ \frac{d^2 J}{dt^2}^{(3)}_{dc,jerk} = \frac{e}{V} \sum_{nk} \left[ 2 \frac{d^n f_n}{dt} \frac{dv_n}{dt}^{(1)} + \frac{d^2 f_n}{dt^2} v_n \right]. \]  

The jerk current’s first and second term in Eq. 4 correspond to the first and second terms of 17, and each represent distinct physical processes. The superscripts \( n \) indicate the \( n \)th-order in the electric fields. The first term represents a process by which carrier accelerate uniformly under a static electric field after being pumped to conduction states. The factors in the first term are

\[ \frac{d^n f_n}{dt} = \omega_m ad E_0^d, \]  

\[ \frac{dv_n}{dt} = \frac{2\pi e^2}{\hbar^2} \sum_c |E_\omega \cdot r_{cv}|^2 \delta(\omega_{cv} - \omega), \]  

\[ \frac{d^2 f_n}{dt^2} = -\frac{2\pi e^2}{\hbar^2} \sum_c |E_\omega \cdot r_{cv}|^2 \delta(\omega_{cv} - \omega), \]  

In the first line we used \( v_n = h\omega_m a d \) and the semiclassical expression \( dk/dt = eE_0/h \) for the evolution of a wave vector under a static electric field. In the second line the expression of the rate of carrier injection into conduction bands is given by Fermi’s golden rule to lowest (second) order. Substituting these into 17 we recover the first term in Eq. 4. One can incorporate phenomenological relaxation times into the above expressions as \(-v_n^0/\tau_m\) and \(-(f_n - f_n^{(0)})/\tau_j\). Addition of these terms gives the basic relaxation already captured by Eq. 3 and a coupling between injection and jerk currents which either vanish or are very small in our case.

‘Doppler’ contribution to jerk current

The second term in Eq. 17 (or Eq. 4) represents a process by which the carrier injection rate itself changes in time. Since the wave vector of a wave packet evolves in time as \( eE_0 t/h \) in the presence of the static field, time-reversed states \( \pm k \) are shifted to \( \pm k + eE_0 t/h \) in the frame of reference where a wave packet is at rest. In this frame clearly the symmetry between \( \pm k \) states is lifted. Alternatively, taking a derivative of Eq. 19 gives

\[ \frac{d^2 f_c}{dt^2} = \frac{2\pi e^2}{\hbar^2} \sum_v \frac{\partial (r_{cv} b_{uc})}{\partial k t} E_0^a E_v^b E_0^d \delta(\omega_{cv} - \omega), \]  

from where the degeneracy between \( \pm k \) states is lifted in the sense that

\[ \frac{d^2 f_n(-k)}{dt^2} \neq \frac{d^2 f_n(k)}{dt^2}, \]  

giving rise to a net current. Note that this process is very different from the injection current which can be derived simply from taking a time derivative of Eq. 16 and keeping only terms contributing to dc current to second order in the optical field

\[ \frac{d}{dt} J^{(2)}_{dc, inj} = \frac{e}{V} \sum_{nk} \frac{df_n}{dt} v_n. \]  

One can check that the injection rates at \( \pm k \)

\[ \frac{d}{dt} f_c(-k) \neq \frac{d}{dt} f_c(k), \]  

are asymmetric only for complex optical field, e.g. with circular or elliptic polarization and hence injection current is nonvanishing only in this case. The jerk current, on the other hand, can be finite for both circular and for linearly polarized optical fields. For times longer than the period of the optical field \( T \) but smaller than \( \tau, \tau_3 \) the current is

\[ J^{(3)}_{jerk} = 6\tau^2 \delta_{abcd} (0,\omega,-\omega,0) E_0^a E_v^b E_0^d. \]

In particular, for a pulse envelop of width larger than \( T \), the time dependance of the current follows the pulse envelope.

Numerical methods

We use density functional theory (DFT) as implemented in the ABINIT [51] computer package, with the generalized gradient approximation to the exchange correlation energy functional as implemented by Perdew, Burke and Ernzerhof.[52] Hartwigs-Goelecker-Hutter norm conserving pseudo potentials [53] were employed. To expand the plane waves basis set, energy cutoffs of

[51] P. E. Blöchl, O. Jepsen, and O. K. Andersen, Phys. Rev. B 49, 16223 (1994).
TABLE I. Peak values of $|\epsilon_3|$ in representative semiconductors obtained from DFT. The photon energy, direct experimental gap, and spontaneous polarization $|\mathbf{P}_0|=P_0$ are indicated. For two-dimensional (2D) materials the effective, i.e., bulk equivalent value is reported.

| Material     | $|\epsilon_3|$ $(\times 10^{-4}$ mA/V$^2$s$^2$) | $\hbar \omega$ (eV) | Direct gap (eV) | $P_0$ ($\mu$C/cm$^2$) | Inversion symmetry |
|--------------|-----------------------------------------------|---------------------|-----------------|------------------------|--------------------|
| Monolayer SnSe | 70                                             | 3.2                | 0.95            | 0.72                   | x                  |
| Monolayer GeS  | 60                                             | 4.5                | 1.89            | 1.95                   | x                  |
| Monolayer GeSe | 55                                             | 3                  | 1.16            | 1.38                   | x                  |
| Monolayer SnS  | 50                                             | 3.5                | 1.57            | 0.95                   | x                  |
| Si            | 20                                             | 3.5                | 3.4             | 0                      | ✓                  |
| GaAs          | 6                                              | 3                  | 1.42            | 0                     | ✗                  |

50 Hartree were employed for GaAs, Si and monolayer GeS and GeSe, and 60 Hartree for SnS and SnSe. We choose the plane of the slab to define the $xy$-plane with the $x$-axis along the spontaneous polarization. The lattice parameter in the $z$-direction is set to 15 Å, which makes for more than 10 Å of vacuum between slabs. To calculate $\epsilon_3$, we included 20 valence and 30 conduction bands for GaAs, Si and GeS and SnS, and 30 valence and 20 conduction bands for GeSe and SnSe. They account for all allowed transitions up to 6 eV.

To extract the effective response of a single layer, we scale the numerical result by the factor $L/d$, where $L$ is the supercell lattice parameter perpendicular to the slab, and $d$ is the effective thickness of the monolayer. For concreteness, we estimate the slab thicknesses as 2.56, 2.59, 2.85 and 2.76 Å for GeS, GeSe, SnS, and SnSe, respectively. Once the ground-state wave function and energies were computed, the TINIBA package [54] was used to compute $\epsilon_3$ as implemented in Ref. [37, 38]. The sum over k-points is made using the interpolation tetrahedron method [55].

**Jerk current tensor in GaAs**

GaAs point group, $43m$, allows three independent tensor components $\epsilon_3^{abcd}$ shown in Fig. 5 as a function of incoming photon energy (frequency).

**Joint density of states**

It is interesting to compare the spectrum of jerk tensor with that of the imaginary dielectric function which follows roughly the joint density of states (JDOS). The imaginary part of the linear dielectric function is given by

$$
\varepsilon_2^{ab} \omega = \frac{e^2 \pi}{\epsilon_0 \hbar V} \sum_{nmk} f_{nm} \rho_{nm}^a r_{mn}^b \delta (\omega_{mn} - \omega),
$$

(25)

The JDOS peaks at points in the BZ which have a high density of states. Four of these points labeled by $E_0$, $E_1$, $E_0'$ and $E_2$ in Fig. 5(b). Comparison with the jerk current allows to investigate the influence of the density of states on the spectrum of $\epsilon_3$. As can be seen, the peak at 3 eV in all of the components of $\epsilon_3$ coincides with $E_1$ indicating a strong correspondence. The peak at 4.5 eV does not exactly correspond to a high density of states. $E_2$ is seen to have only a small kink effect on the $xxxyy$ component.

**Spin-orbit coupling**

The spin-orbit coupling is known to be important in materials that break inversion symmetry such as GaAs. In GaAs, the SOC field can be large near the band edge. Hence we expect corrections to the jerk tensor spectrum from the SOC. In Fig. 6(a) and (b) we show the $xyyx$ and $xxxy$ components of the $\epsilon_3$ with and without SOC. In both components the SOC produces a shift towards lower energies and is most noticeable near the points of high JDOS. The longitudinal component $xxxy$ increases $\times 5$ near 4.7 eV whereas the transverse component only increases by a fraction. For both components the direction of the current is mostly unchanged. Interestingly, the contributions from SOC in the jerk spectrum of Si are also large $\times 5$ even though Si does not break inversion symmetry.

**Doppler vs classical terms**

In Fig. 7 we present the contributions from the first and second terms to $\epsilon_3$ (SOC included). Interestingly, the quantum contribution is larger relative to the classical contribution in the transverse component $xxxy$ and weaker in the longitudinal components $xyyx$ and $xxxx$. However, the sign of the quantum term is opposite to the sign of the classical acceleration leading to a partial cancellation and a smaller transverse component. There seems to be a strong correlation between the sign of these two contributions in the transverse components of all materials studied.
Jerk current in Si

Si point group, $m3m$, and allows three independent tensor components $xyyx$, $xxyy$, and $xxxx$ shown in Fig. 5 as a function of incoming photon energy. The jerk spectrum vanishes for incoming photon energies less than the (direct) energy band gap of 3.5 eV. It exhibits similar characteristic isolater peaks as for GaAs. The spectrum peaks at the band edge and at 5.4 eV, both peaks of the order of $24 \times 10^{14}$ mA/V$^3$s$^2$ and $15 \times 10^{14}$ mA/V$^3$s$^2$ respectively. Note that current transverse to the static field (Hall current) is possible and is controlled by the component $xxyy$ of $\iota_3$, but it is an order of magnitude smaller with respect to the longitudinal components. Similar to GaAs, peaks at 3.5 eV and 5.4 eV in two of the the components of $\iota_3$ coincides with a peaks in the JDOS. Similar to GaAs including SOC enhances the response up to a factor of 3.

Interestingly, the doppler contribution to jerk current is larger relative to the classical contribution in the transverse component $xyyx$ and weaker in the longitudinal components $xyxy$ and $xxxx$. However, the sign of the doppler term is opposite to the sign of the classical acceleration leading to a partial cancellation and a smaller transverse component. As we see there seems to be a strong correlation between the signs of these two contributions in all the transverse components of all materials studied.
FIG. 7. Classical and Doppler contributions to \( \iota_3 \) in GaAs, see Eq. 17.

**Jerk current tensor in monolayer GeS, GeSe, SnS and SnSe**

Monolayer GeS, GeSe, SnS and SnSe are predicted to have large in-plane spontaneous polarization, see Table I. Let us chose the slab to define the \( xy \)-plane with the \( x \)-axis along the polarization axis and \( z \) out of the slab. The point group of monolayer GeS is \( mm2 \) and accordingly, there are six independent components \( yyyy \), \( xxxx \), \( yxx \), \( yyy \), \( xxx \), and \( xyx \) shown in Fig. 8 as a function of photon energy. The 2D values are reported as bulk equivalent, i.e., per unit volume. This is to be able to compare easily with GaAs and Si. Out of plane response is much weaker and is not considered here.

The spectrum of \( \iota_3 \) in monolayer GeS GeSe, SnS and SnSe is flatter than for GaAs and Si which are composed of isolated peaks corresponding to high JDOS. The average could be \( 20 \times 10^{14} \text{ mA/V}^3\text{s}^2 \) (or more) for longitudinal components \( yyyy \), \( yxx \) in a broad range of photon energies 2.7-5.5 eV. The range of frequencies includes a part of the visible spectrum. The transverse components \( yyx \) and \( xyx \) are an order of magnitude smaller than the longitudinal components. Importantly the magnitude of \( \iota_3 \) is generally an order of magnitude larger than of bulk GaAs and Si. This is more evident in monolayer SnS where \( \iota_3 \) can reach peak values of \( 70 \times 10^{14} \text{ mA/V}^3\text{s}^2 \). Note the large response perpendicular to the polarization (yyyy) with respect to the response along the polar axis (xxxx). This is explained by the partial cancellation along the polar axis between the classical and Doppler terms Eq. 17 which have opposite signs, see Fig. 10. Note also that the current perpendicular to the polarization described by \( yyyy \) and \( yxx \) is very sensitive to the direction of the optical field, \( y \) and \( x \), respectively. A change in the polarization of the optical field from \( x \) to \( y \) can even changes the sign of the photocurrent. This is not the case for the current along the polar axis described by \( xxxx \), \( xyx \) where a change in optical field polarization from \( x \) to \( y \) will only decrease/increase the current.

**Joint density of states**

The JDOS of GeS (and related materials) is uniform over a large frequency range with small fluctuations near points of high JDOS, see Fig. 8. The overall magnitude of the JDOS of monolayer GeS is at most \( \times 2 \) that of GaAs which suggests the absolute magnitude of the density of states is not the origin of the \( \times 10 \) difference \( \iota_3 \) in these materials. In monolayer GeS, of the peaks in the JDOS coincide with those of the jerk current spectrum but it is rather difficult to find exact coincidences.

**Spin-orbit coupling**

The SOC in monolayer GeS (and related materials) is small, of the order of \( \sim 20 \text{ mV} \), yet SOC increases the jerk spectrum by \( \times 20 \) with respect to the case with no SOC for all longitudinal components. Fig. 9 shows the comparison of the jerk current spectrum for the largest components of monolayer GeS, \( yyyy \), \( yxx \), and a transverse component \( yxx \). The transverse component do not change significantly with addition of SOC. GaAs also shows an enhanced response of the longitudinal components with SOC but only by \( \times 2 \), which suggests reduced
dimensionality plays a big role in the large response.

**Doppler vs classical terms**

Fig. 10 shows the classical (first) and Doppler (second) contributions to \( \epsilon_3 \) of monolayer GeS. Note that the 1st and 2nd contributions have opposite signs, except for the component perpendicular to the polar axis \( yyyy \).
relatively insensitive to the direction of the optical field. There is a partial cancellation of between the 2nd and 1st contributions in the longitudinal $xxx$ and transverse $yyxx$, $xxyy$ components and a constructive interference in the $yyyy$ component. We see that the current along the polarization axis is relatively insensitive to the direction of the optical field whereas current along the perpendicular $y$ axis is very sensitive on the direction of the optical axis. The current along the polar axis is dominated by the polarization and not by the band structure. This behavior is rather different from GaAs where there is no polar axis.