Ballistic and shift currents in the bulk photovoltaic effect theory

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The bulk photovoltaic effect (BPVE) – generation of electric currents by light in noncentrosymmetric materials – was investigated massively in 70th and 80th of the last century. Hundreds of experimental and theoretical papers were published and tens of different materials (ferroelectrics and piezoelectrics) were considered. The results of these studies are summarized in book [1]. They cover many aspects of the BPVE: a general definition of the effect, its mechanisms for different types of light-induced transitions, applications to particular materials, the influence of magnetic field, comparison between theory and experiment, and also delusions occurred. Numerous further applications to semiconductor nanostructures are summarized in book [2].

A new upsurge of interest in the BPVE occurred several years ago. It is caused by the progress in material science, by increased computational possibilities, and by the prospects of employment of the BPVE in efficient light batteries. Unfortunately, the ongoing progress is aggravated by oversights in the basics of the BPVE theory and by misinterpretations of the experimental and theoretical results. These drawbacks are centered around the notion of the so-called shift current. They are relevant to numerous recent publications [3–12]. Our goal is to outline what and why is not correct in the latest developments to return the studies on the right track. When necessary, we refer to original papers cited in [1, 2].

The BPVE is conventionally defined by the tensorial relation for the DC current density $j$:

$$ j_i = (\beta^L_{inm} e_n e^*_m + \beta^C_{inm} \kappa_n) I. $$

(1)

Here $I$ is the light intensity, $e$ is the unit polarization vector, $\kappa = i(e \times e^*)$, and $\beta^L_{inm} = \beta^L_{inm} \kappa_n$ and $\beta^C_{inm} = \beta^C_{inm} \kappa_n$ are two photovoltaic tensors possessing the symmetry of the piezo- and gyration tensors, respectively. This definition employs nothing, but symmetry considerations. The first contribution to $j$ is nonzero for the linear polarization ($e = e^*$); it corresponds to the so-called linear BPVE.

The second contribution is zero for the linear polarization and maximal for the circular polarization ($|\kappa| = 1$); it corresponds to the circular BPVE. Using Eq. (1), numerous experimental data on $j(I, e)$ were identified with the BPVE, and nonzero components of the tensors $\beta^L$ and $\beta^C$ were measured for tens of noncentrosymmetric materials, including ferroelectrics BaTiO$_3$, LiNbO$_3$ and cubic piezoelectric crystals GaAs, GaP.

The microscopic theory includes general relations, simplified models, and applications to particular materials. Most of the results are obtained within the paradigm of quasi-free band electrons implying that the electron (hole) momentum $k$ exceeds the reciprocal of the free-path length $\ell$. The opposite case of hopping transport is also considered. The crystal asymmetry is presumed to be small. This means that the asymmetry parameter $\xi_0$, defined as the ratio of non-central ionic displacement in a unit cell to the cell size $a$, is small, $\xi_0 \ll 1$.

**Ballistic and shift currents:** A cornerstone of the microscopic BPVE theory is that the total current density $j$ is generally the sum of two physically different ballistic and shift currents, $j = j_b + j_{sh}$. The ballistic current is

$$ j_b = -e \sum_{s,k} f_{s,k} v_{s,k}, $$(2)

where $e$ is the elementary charge, $v_{s,k} = \nabla_k \varepsilon_{s,k}/\hbar$ is the electron velocity, and $f_{s,k}$ is the momentum distribution in band $s$. This contribution is due to asymmetry of the momentum distributions, $f_{s,k} \neq f_{s,-k}$. It can be regarded as classical one because of the relevance to an abundance of charge-transport phenomena. The shift current $j_{sh}$ is caused by shifts of electrons during light-induced transitions [1, 2, 13]. It originates from non-diagonal in $s$ elements of the electron-density matrix.

In noncentral media, any electronic process, including photo-excitation, recombination, elastic and inelastic scattering, is asymmetric. Correspondingly, kinetic equations for $f_{s,k}$ are not inversion invariant. In the absence...
of thermal equilibrium, each process contributes to \( j_s \). However, in thermal equilibrium, where the detailed balance between transitions \( s, k \rightarrow s', k' \) and \( s, k \rightarrow s', k' \) takes place, the kinetic equations give \( k \)-symmetric Fermi distributions for electrons and holes and \( j_h = 0 \). The principles of calculation of \( j_h \) are documented for all main types of light-induced transitions including trap-band and band-band transitions \[1, 14, 17\]. They are illustrated with simple models. Existence of \( j_h \) is undeniable for both the linear and circular BPVE.

Difficulties in calculation of \( j_h \) for particular materials are rooted not only in an insufficient knowledge of the band structure. They stem greatly from a bad knowledge of the kinetic characteristics, such as the momentum relaxation times, the energy relaxation times, the recombination times that involve numerous electron and phonon bands and interaction channels. This situation is not special for the BPVE. It is relevant to other light-induced charge transport phenomena, such as, e.g., the photon-drag effect. Nevertheless, there are cases where \( j_h \) is calculated and compared with experiment.

Regardless of details, the value of \( j_h \) for trap-band and band-band transitions can be evaluated as

\[
j_h = e g \xi \ell_0,
\]

where \( g = \alpha I/\hbar \omega \) is the generation rate, \( \alpha \) is the light absorption coefficient, \( \hbar \omega \) is the light-quantum energy, \( \ell_0 = v_0 \tau_0 \), \( v_0 \) is the free-path length of photo-excited hot electrons (holes), \( \tau_0 \) is their momentum relaxation time, and \( \xi \) is the excitation asymmetry parameter. The latter is the only BPVE-specific parameter in Eq. (3). According to model estimates, \( \xi \) ranges from \( 10^{-1} \) to \( 10^{-3} \).

Properties of the electronic wave function \( \Psi_{s, k} (r) \) have to be commented to avoid confusion. One might suggest that it is a Bloch function \( \Psi_{s, k}^B (r) \) obeying the micro-reversibility relation \( \Psi_{s, k} = \Psi_{s, -k}^B \). With this choice, the excitation and recombination rates for electrons are symmetric in \( k \) for the linear light polarization and \( j^B_{h} = 0 \). However, identification of \( \Psi_{s, k} \) with \( \Psi_{s, k}^B \) is generally incorrect. For trap-band transitions, the effect of trap potential is important. Here, the functions \( \Psi_{s, k}^B (r) \) and \( \Psi_{s, k}^B (r) \) comprising converging (−) and diverging (+) waves, see Fig. 1a, must be used to describe the excitation and recombination probabilities \[3\]. None of the \( \Psi_{s, k}^B \) functions obeys the micro-reversibility relation; this gives asymmetry of the excitation and recombination rates. At the same time, the micro-reversibility relation \( \Psi_{s, k}^B = \Psi_{s, -k}^B \) links the differential probabilities of excitation and recombination: \( u_{s, k}^{ex} (e) = u_{s, -k}^{rec} (e) \). An analogous situation takes place for VB→CB transitions. The Coulomb interaction of light-induced electrons and holes, which is typically strong, causes not only localized excitonic states, but also inequality \( \Psi_{s, k} \neq \Psi_{s, -k}^B \) \[3, 17\].

The terms shift BPVE and \( j_{sh} \) were introduced in 1982 at the breakthrough in understanding of the physics of the non-diagonal contribution to \( j \) \[13\]. It was realized that an electronic transition from Bloch state \( \left| n \right> = \left| s, k \right> \) to Bloch state \( \left| n' \right> = \left| s', k' \right> \) is accompanied by the shift \( R_{n' n} \) within a unit crystal cell:

\[
R_{n' n} = -R_{n n'} = -(\nabla_k + \nabla_{k'}) \Phi_{n' n} + \Omega_{n} - \Omega_{n'},
\]

where \( \Phi_{n' n} \) is the phase of the transition matrix element and \( \Omega_n \) is the Berry connection as defined in \[10\]. The shift is even in \( k \) and \( k' \) and nonzero only in the absence of inversion symmetry. It does not depend on the choice of the phase of the \( \Psi_{s, k}^B \) (the gauge invariance). The shift current is expressed generally by \( R_{n' n} \) as \[12, 13\]

\[
j_{sh} = e \sum_{n, n'} W_{n' n} R_{n' n},
\]

where \( W_{n' n} \) is the transition rate from \( n \) to \( n' \). In thermal equilibrium, where the detailed balance takes place, \( j_{sh} = 0 \). Since \( R_{n' n} \) already accounts for the crystal asymmetry, \( j_{sh} \) has to be calculated on symmetric distributions, \( f_{s, k} = f_{s, -k} \). Small differences between \( \Psi_{s, k}^B \) and \( \Psi_{s, k} \) are not important for the shift BPVE.

Application of Eqs. (4) to light-induced band-band transitions gives an explicit expression for \( j_{sh} \) \[12, 13\]. It consists of partially compensating contributions \( j_{sh}^{rec} \) and \( j_{sh}^{rec} \) relevant to electron shifts during the excitation and recombination processes. The excitation contribution \( j_{sh}^{rec} \) is proportional to the light intensity \( I \) and includes no kinetic characteristics. The recombination contribution \( j_{sh}^{rec} \) incorporates such characteristics: Hot electrons (holes) experience the energy relaxation and recombine being predominantly (or partially) thermalized, see Fig. 1b. Since the rates of excitation and recombination are the same in steady state, the value of \( j_{sh} \) depends on particular values of the shift for hot and thermalized electrons. In the absence of the relaxation, as in the case of Fig. 1c, the total shift current would be zero because of an exact compensation of \( j_{sh}^{rec} \) and \( j_{sh}^{rec} \). Simple models show that the contribution \( j_{sh}^{rec} \) can be dominating in
ferroelectrics \cite{1,20}. The value of the shift current can be estimated as
\[ j_{sh} = e g \bar{R}, \]
where \( \bar{R} \approx \xi_0 a \) is an effective shift. The condition of dominating ballistic current is thus \( \xi_{ex} l_0 \gg \bar{R} \). It is expected to be fulfilled for VB→CB transitions where the strong electron-hole interaction provides the excitation asymmetry parameter \( \xi_{ex} \approx \xi_0 \). In the case of transition between electronic (or hole) bands we have \( \xi_{ex} \approx a/l_0 \) and \( j_b \approx j_{sh} \). More accurate calculations support these simple estimates, see also below.

An alternative BPVE theory: Works \cite{2,4,21} put forward an alternative version of the linear BPVE theory which strongly contradicts to \cite{1,2}. This version is centered around the notion of shift current and deals with band-band transitions. It treats the linear BPVE as a dynamic nonlinear-optical phenomenon, free of influence of kinetic parameters. The classical ballistic contribution \( j_b \), caused by asymmetry of the momentum distributions for electrons and holes, is absent within this approach. The basic relation for \( j_{sh} \) is different here from the expression of \cite{1,13} for \( j_{sh}^{ex} \). In other words, the shift currents considered do not include the recombination contribution \( j_{sh}^{rec} \). Since the expression for \( j_{sh}^{ex} \) includes only band (but not kinetic) characteristics, it can be used for calculations from "first principles". Such calculations have been performed for a number of ferroelectric materials \cite{3,5,12}. Broad frequency ranges, which are not restricted to the excitation of charge carriers near band edges, is a remarkable feature of these calculations. Neither discussion nor criticism of \cite{1} can be found within the alternative BPVE theory.

Remarkably, the formulae for \( j_b \) of \cite{21,22} served as the basis for the new developments. The model employed in these papers includes only the interaction of Bloch electrons with a classical electromagnetic field. The electron-phonon and electron-hole interactions leading to the ballistic current \( j_b \), as well as the common processes of energy relaxation, thermalization, and recombination, are beyond this oversimplified model. The authors of \cite{22} and of the computational papers \cite{3,5,12} refer to \cite{1} as to a general source, but pay no attention either to the specific results on \( j_b \) and \( j_{sh} \) or the general argumentation. In essence, the model expression for \( j_b \) of \cite{21,22} is considered as omnipotent one for the linear BPVE. Referring to \cite{3,4,21,22}, numerous experimental papers \cite{23,24} claim for observation and employment of the shift currents.

Surprisingly, no attention was paid to numerous evidences (qualitative, quantitative, experimental) of the presence of the ballistic contributions to \( j_b \) as well as to pathology of the model ignoring the energy relaxation and recombination processes. Ignorance of the recombination leads evidently to a wrong conclusion about the presence of steady-state shift current for an ensemble of localized two-level centers subjected to the resonant excitation, see Fig. 1c. For crystals of the pyroelectric symmetry, it leads to nonzero currents in thermal equilibrium owing to the thermal-photon excitation processes, i.e., to perpetual motion machine of the second kind.

Applications to GaAs crystals: The BPVE theory was applied to a number of noncentrosymmetric semiconductors with well known band structure and explored kinetic properties (GaAs, GaP, Te). The results of the calculations, performed for near-band-edge absorption, have showed a good agreement with experimental data \cite{1}. Altogether, they evidence that the ballistic current \( j_b \) is either dominating over or comparable with \( j_{sh} \). This refers both to the linear and circular BPVE.

Here we exhibit some results obtained for GaAs. This cubic piezoelectric possesses only the linear BPVE characterized by the component \( \beta = \beta_{23} \) of the photovoltaic tensor \( \beta^L \). The band structure of GaAs for \( ka \ll 1 \) is shown in Fig. 1a. It is characterized by a single CB and a compound VB comprising the light (\( h_l \)), heavy (\( h_h \)), and split-off (\( h_\Delta \)) hole sub-bands. The bandgap \( E_g \) and spin-orbit splitting \( \Delta \) are \( \approx 1.53 \) and 0.34 eV, respectively. The band structure is described by the Kane Hamiltonian which is a \( k \)-dependent \( 8 \times 8 \) matrix \cite{30}. Calculations of \( j_b(\omega) \) for CB→VB transitions were performed in \cite{31,53} near the fundamental absorption edge, \( E_g < h\omega < E_g + \Delta \) for He-temperatures. The Coulomb interaction between photo-excited electrons and holes was found to be the dominating mechanism of the excitation asymmetry. Complexity of the quantitative calculations is rooted not only in simultaneous excitation of three bands, but also in involvement of different mechanisms of momentum relaxation. The strongest of them is scattering on optical phonons. This mechanism is allowed for sufficiently large distances \( h\omega - E_g \) and gives only partial momentum relaxation. This is why weaker kinetic processes – scattering on acoustic phonons and remnant neutral and charged point defects – contribute to the relaxation. As the result, the dependence \( \beta(h\omega) \) acquires a characteristic jagged structure, see Fig. 2b, which is the fingerprint of the ballistic nature of \( j \). The shift current \( j_{sh} \) was found to be smaller than \( j_b \) by more

![FIG. 2: a) Band structure of GaAs and the main light-excitation channels (blue arrows). b) Spectral dependence of the BPVE-tensor element \( \beta \) and experiment \cite{1,53}](image-url)
than one order of magnitude.

BPVE experiments were performed with epitaxial films of GaAs at $T = 4.2$ K \[33\]; they included also Hall measurements. The main features of the experimental spectrum in Fig. 2b nicely correspond to the theory (to the spectral dependence of the free-pass length $l_0$). The maximal values of the Hall mobility and free-pass length occur at $\approx 1.56$ eV; they are about $5 \times 10^5$ cm$^2$/Vs and 8 $\mu$m. The corresponding excitation-asymmetry parameter is $\epsilon_{ex} \approx 2 \times 10^{-3}$.

Importantly, theoretical and experimental studies of two allied kinetic photovoltaic phenomena – the photodrag effect and the surface photovoltaic effect – were performed independently for the same interband transitions in GaAs at Helium temperature $\approx 300$ K. \[34, 35\]. A good agreement between complicated theoretical and experimental spectral dependences was obtained. A great body of results gives thus a comprehensive knowledge of the interband photovoltaic effects in GaAs.

Relationship between $j_h$ and $j_{sh}$ changes dramatically for transitions between the $h_l$ and $h_h$ hole sub-bands when the Coulomb mechanism of the ballistic current is absent. In particular, it was shown theoretically and experimentally for $h\omega = 117$ meV that a strong compensation of $j_h$ and $j_{sh}$ takes place in the temperature range $(130 - 570)$ K. \[36, 37\]. The total current $j = j_h + j_{sh}$ is relatively small and sign-changing.

**Influence of magnetic field:** Investigation of the influence of magnetic field on the photovoltaic current sheds light on the nature of the shift currents and on close links between $j_h$ and $j_{sh}$. Since the electronic shifts occur instantaneously during the light-induced transitions, one might suggest that the effect of magnetic field $H$ on $j_{sh}$ is negligible compared to the effect on $j_h$. The latter can be viewed for small fields as generation of the Hall current $\delta j_h \approx (\mu_0/c)(H \times j_h)$, where $\mu_0$ is the mobility of the dominating photo-excited charge carriers, $c$ is the speed of light, and the signs $+$ and $-$ correspond to dominating electrons and holes. This effect is due to the action of the Lorentz force during the free electron path. Applying large magnetic fields, $\mu_0 H/c \gg 1$, leading to suppression of the transverse (to $H$) ballistic current, one can hope to get experimentally the transverse shift current.

Unfortunately, the above suggestion on a negligible influence of the magnetic field on $j_{sh}$ is not fully correct. The point is that this field breaks the time-reversal invariance, so that $\Psi^B_{k+} \neq \Psi^B_{-k}$ for Bloch electrons. This causes magneto-induced asymmetry of photo-excitation (the inequality $w^{ex}_k \neq w^{ex}_{-k}$) without any perturbing effect of the electron-hole and electron-phonon interactions. Correspondingly, an additional magneto-induced contribution $\delta j^M_{sh}$ to the ballistic current joins the game. Surprisingly, this ballistic contribution is expressed by the shift current $\delta j^M_{sh} = \pm (C\mu_0/c)(H \times j_{sh})$, where $C$ is a model-dependent dimensionless constant whose value can be smaller or modestly larger than 1. This formula shows that the shift and ballistic currents become mutually related in the presence of magnetic field. For the large magnetic fields, the current $\delta j^M_{sh}$ compensates the transverse component of the shift current, so that the total transverse current vanishes. Thus, a decisive phenomenological separation of $j_b$ and $j_{sh}$ is not possible in Hall experiments. With simplifying assumptions, one can demonstrate merely that $j_{sh}$ is present.

Employment of the Hall model for determination of the mobility and sign of the photo-excited electrons is justified only in the cases when $j_h \gg j_{sh}$, including the VB→CB and trap-band transitions. Large values of the mobility obtained in such experiments \[31, 33, 40\], $\mu_0 \gtrsim 10^5$ cm$^2$/Vs, support once again the conclusion about smallness of the shift current and evidence in favor of the ballistic models of the BPVE.

**Summary:** There are no reasons to identify the measurable total BPVE current $j$ or the linear BPVE current $j^l$ with the shift current $j_{sh}$. Numerous theoretical and experimental arguments indicate that the classical ballistic contribution $j_b$, caused by asymmetry of the momentum distributions for electrons and/or holes, is either dominating or substantial. The ballistic current is missed in within the alternative BPVE theory. The recombination contribution $j_{sh}^{rec}$ to the shift current, which is nonzero in ferroelectric crystals, is also missed within the alternative theory. The latter leads to perpetual motion machine of the second kind.

Both missed contributions, $j_b$ and $j_{sh}^{rec}$, involve kinetic characteristics, such as momentum and energy relaxation times. Correspondingly, it is necessary to treat the BPVE as a kinetic phenomenon, substantially different from and more complicated than the quadratic-response effects caused solely by the band structure. The difference stems from the fact that generation of a DC current is always a dissipative process in contrast to generation of the nonlinear polarization. This difference disappears in the frequency domain when the frequency difference between two light waves substantially exceeds the reciprocal of the momentum relaxation time $\tau_0$.

The shift and ballistic contributions to the BPVE current possess the same symmetry properties, they cannot be decisively separated in light-polarization experiments. The same is valid greatly with respect to Hall measurements. Deep knowledge of the electronic band structure and kinetic processes is necessary to judge about the relationship between $j_b$ and $j_{sh}$.

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