

Simulations of ultrafast laser-induced excitation and heating of electron sub-system of semiconductors with the Vinogradov equation and multi-band Keldysh formula

Vitaly Gruzdev^{*(1)}, Drake Austin⁽²⁾, Olga Sergaeva⁽¹⁾, Enam Chowdhury⁽¹⁾

(1) Department of Mechanical and Aerospace Engineering, University of Missouri, Columbia, MO, 65211, USA

(2) Department of Physics, The Ohio State University, Columbus, OH, 43210, USA

*Contact author: gruzdevv@missouri.edu

Abstract

High-intensity laser interactions with band-gap solids are frequently simulated with the Keldysh photoionization-rate formula and the Drude model to describe light absorption and related heating of conduction-band electrons. That approach suffers from several internal contradictions. We fix some of them by combining the Vinogradov equation for energy absorption with multi-band version of the Keldysh equation for the rate of photoionization.

1. Introduction

Recently wide-gap semiconductors attracted significant attention due to the expansion of their applications in photonics [1] and studies of strong-field ultrafast laser effects in solids [2]. Many of the applications consider interaction of the semiconductors with high-power ultrashort (femtosecond) laser pulses either for processing or for device operation. The first step of the interaction is a deposition of laser-radiation energy in electron subsystem that has two major contributions [3-5]. The first one is done by electron excitation from valence to conduction band (by the photo- or impact ionization). The other one is by intra-band absorption by the laser-generated conduction-band electrons via electron-phonon collisions [2-5]. The photoionization and associated nonlinear absorption of laser radiation are usually described by the Keldysh formula [6] under the two-band approximation (one valence band and one conduction band) [2-5]. Laser-driven collisions of the conduction-band electrons and their contributions to optical response and energy absorption are frequently simulated by the Drude model [2-5]. That approach is applied to the ultrafast laser-semiconductor interactions in a broad range of laser and material parameters. However, deeper analysis [7, 8] shows that the Drude approximation is not valid at laser intensity characteristic of initiating of the high-power laser-induced interactions, e. g., ablation. Another significant gap of that traditional Keldysh-Drude approach includes the neglected contribution of multiple valence bands into multiphoton transitions (Fig. 1) Finally, the laser-driven oscillations and corresponding pondermotive energy of the conduction electrons are neglected by the Drude model [2-5], but they are taken into account by the Keldysh formula combined with the

Drude model [2-5]. Recent results from ultrashort pulse laser damage experiments in mid-IR wavelengths seem to accentuate the discrepancy even further [9], as pondermotive energy scales as λ^2 . To fix those contradictions and gaps, we report here a new approach to simulate laser-driven inter-band electron excitation, and linear and non-linear energy absorption in wide-band-gap semiconductors with direct-gap band structure.

2. Multi-band Keldysh-Vinogradov model

For simplicity, a direct-gap cubic crystal is considered below with electric field of laser radiation directed along one of principle crystal axes. Parameters of ZnSe [10] are utilized for the simulations reported here. The proposed model considers inter-band electron transitions from heavy-hole (HH), light-hole (LH), and split-off (SO) valence bands (Fig. 1). Energy-momentum relations are parabolic for the conduction and the SO bands, but they are the Kane-type relations [6] for the HH and LH bands that is characteristic of typical semiconductor [10]. Since the energy gap between the higher valence bands and the split-off band is about $10^2 - 10^1$ eV at the Γ point [10], simulations of the photo-ionization rate with the Keldysh formula [3] show very similar contributions of all the three valence bands to the total photoionization at high intensity (Fig. 2). Following that result, time variations of

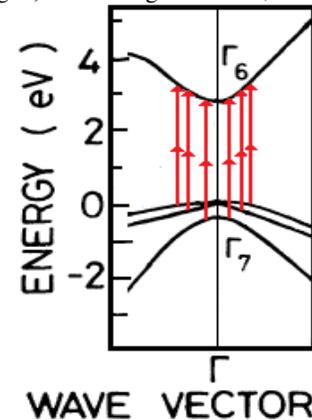


Figure 1. The band structure of a typical wide-gap semiconductor and possible two-photon inter-band transitions (red arrows) to the lowest conduction band from heavy-hole (HH), light-hole (LH), and split-off (SO) valence bands around Γ -point of the Brillouin zone.

conduction-band electron density is simulated by the following rate equation that includes the contributions from the three inter-band transitions:

$$\frac{dN_{CB}}{dt} = f_{HH}(t) \cdot W_{HH}[I(t)] + f_{LH}(t) \cdot W_{LH}[I(t)] + f_{SO}(t) \cdot W_{SO}[I(t)] \quad (1)$$

where the factors f_{HH} , f_{LH} , and f_{SO} describe dependence of the corresponding inter-band transitions on population of the involved bands. W_{HH} , W_{LH} , and W_{SO} are the rates of inter-band electron up-transitions from HH, LH, and SO valence bands evaluated by the Keldysh formula for HH and LH bands, and by the parabolic-band modification of the Keldysh formula for the SO band. The inter-band transitions are treated as independent within the Keldysh approach [6] as long as the ionization rates are not very high. Rate of energy absorption contains contributions from the photo-ionization evaluated utilizing effective (i.e., laser-modified) band gaps suggested by the Keldysh model [6]:

$$\begin{aligned} \frac{dE}{dt} = & f_{HH}(t) \cdot W_{HH}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{HH}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega \\ & + f_{LH}(t) \cdot W_{LH}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{LH}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega \\ & + f_{SO}(t) \cdot W_{SO}[I(t)] \cdot \left\langle \frac{\Delta_{ef}^{SO}[I(t)]}{\hbar\omega} \right\rangle \cdot \hbar\omega + \left(\frac{d\varepsilon}{dt} \right)_{CB} \end{aligned} \quad (2)$$

where angle brackets denote taking an integer number of laser photons required to bridge corresponding effective band gaps [6].

The last term of Eq. (2) is the contribution of the conduction-band electrons absorbing light via electron-phonon collisions. It is simulated by the Vinogradov equation for energy-absorption rate by a single conduction electron [7]:

$$\begin{aligned} \frac{d\varepsilon}{dt} = & \left(\frac{eF(t)}{\omega} \right)^3 \frac{2G^2 k_B T_{ph}}{\pi^2 \hbar^4 \rho u^2} \Psi_1(\gamma) \\ & + \left(\frac{eF(t)}{\omega} \right) \frac{e^2 \Omega_0 (\varepsilon_0 - \varepsilon_\infty)}{\pi \hbar \varepsilon_0 \varepsilon_\infty} \coth \left(\frac{\hbar \Omega_0}{2k_B T_{ph}} \right) \Psi_2(\gamma) \end{aligned} \quad (3)$$

where the functions Ψ_1 and Ψ_2 are defined in Ref. [7], and the Keldysh parameter γ is introduced as usually [6]:

$$\gamma = \frac{\omega \sqrt{2m\varepsilon(t)}}{eF(t)} \quad (4)$$

The conduction-electron absorption rate of the right-hand part of Eq. (3) is depicted in Fig. 3. We note that laser parameters characteristic of high-intensity laser-semiconductor interactions meet the requirements of the classical approximation for the Boltzmann kinetic equation that underlies the Vinogradov equation [7, 8]. Low rate of light absorption by the conduction-band electrons suggests that impact ionization makes a negligible contribution as compared to the multi-band photoionization.

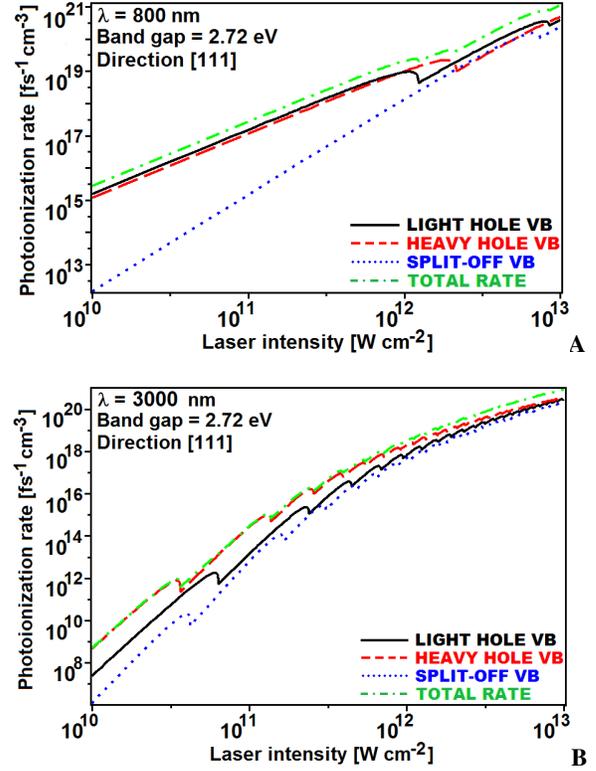


Figure 2. The photo-ionization rates of Eq. (1) evaluated by the Keldysh formula for electron transitions from heavy-hole, light-hole, and split-off valence bands to the lowest conduction band of ZnSe (Γ -point; electric field is parallel to [111] direction of the crystal) at wavelength 800 nm (A) and 3000 nm (B).

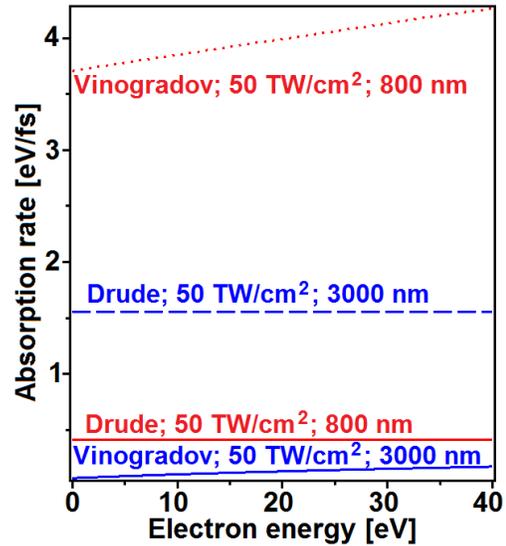


Figure 3. Rate of energy absorption by single conduction-band electron from the Vinogradov equation Eq. (3) [7] evaluated as a function of electron energy for laser intensity 50 TW/cm², material parameters of ZnSe [10], and laser wavelengths 800 and 3000 nm. Single-electron absorption rate by the Drude model is depicted for similar parameters by assuming 1 fs electron-photon-phonon collision time.

The procedure of numerical evaluation of the rate of light absorption by conduction-band electrons from the Vinogradov equation [7] delivers energy distribution of the conduction-band electrons. An example of the distribution is depicted in Fig. 4 for laser wavelength 3600 nm, pulse width 90 fs, and peak laser fluence 1.01 J/cm². Generation of significant high-energy electron density is attributed to large pondermotive energy characteristic of electron oscillations driven by longer wavelengths of laser radiation. Specific structure of the energy distribution results from the contributions of the three valence bands.

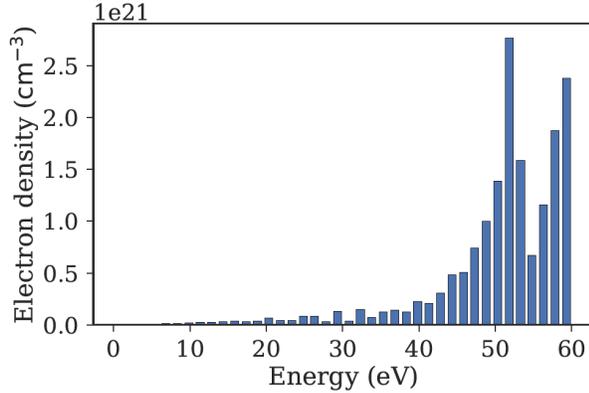


Figure 4. Energy distribution of electrons within the conduction band (zero energy corresponds to the bottom of the conduction band) simulated with Eq. (3) at the end of a single 90-fs laser pulse at central wavelength 3000 nm and peak fluence 1.01 J/cm². Laser-induced variations of real part of refractive index were neglected for simulations of this energy distribution.

3. Discussion

Three upper valence bands can make very similar contributions to the total photoionization rate in semiconductors (Fig. 1). The two-band approximation can be acceptable at low laser intensity only at specific wavelengths because the HH and LH contributions to the multiphoton ionization are similar if energy of a laser photon is 30-50% of the band gap energy. Therefore, accounting for multiple bands is required to properly evaluate the photoionization rate.

The Vinogradov model allows accounting for band-structure modification by pondermotive potential of laser-driven electron oscillations for both the photoionization inter-band transitions and the intra-band absorption by the conduction-band electrons. The rate of energy absorption by conduction electrons evaluated from the Vinogradov model [7] deviates from the estimations of the Drude model by as much as factor of 10. We also note that the deviation becomes more pronounced with increase of laser wavelength in spite of that the total intra-band absorption reduces due to reduction of single-photon energy. Moreover, the Vinogradov model predicts higher absorption by conduction electrons at shorter wavelengths

while it predicts lower absorption at longer wavelength compared to the estimations from the Drude model.

Energy distribution of conduction-band electrons demonstrates specific structure associated with individual contributions of each participating valence band. Also, significant promotion of conduction-band electrons to higher energy levels by long-wavelength laser radiation (Fig. 4) is justified by the fact that the longer wavelength, the larger the pondermotive energy is transferred to oscillating electrons. This effect can hardly be expected from the simulations that neglect the electron oscillations in the conduction band.

Therefore, the contribution of pondermotive energy of oscillating conduction electrons should not be neglected in evaluation of the absorption rate by conduction-band electrons. Moreover, the Vinogradov model introduces dependence of the conduction-electron absorption on electron energy that is completely neglected by the traditional Drude model [3-5, 11, 12]. Those advantages suggest considering the multi-band Keldysh-Vinogradov model as a more advanced and solid theoretical basis for simulations of the ultra-fast laser-semiconductor interactions.

We also note that the simple parabolic and Kane-type relations utilized for this approach become invalid and inaccurate with increase of the pondermotive energy of oscillating conduction electrons due to increase of laser wavelength. This issue results from the general drawback of those simplified band-structure models: they do not properly describe Bragg-type electron reflections of oscillating electrons at edges of Brillouin zone [12-14]. Therefore, one of the most reasonable improvements of this approach considers utilizing of more advanced approximations for energy bands, e.g., cosine relations that provide correct treatment of the Bragg reflections of electrons oscillating along one of principle axes of a crystal [13, 14].

4. Conclusions

In summary, we propose multi-band Keldysh-Vinogradov model as a more reasonable approximation for modeling of laser-driven electron excitation and associated optical response. It fixes the gaps, contradictions, and issues of the traditional combination of the Drude and Keldysh models [3-5] by accounting for modification of energy of conduction-band electrons by the pondermotive potential of laser-driven Bloch oscillations. The proposed approach is essentially simpler and faster for implementation than a direct numerical solving the Boltzmann equation.

5. Acknowledgements

This material is based upon work supported by the Air Force Office of Scientific Research under award numbers FA9550-16-1-0069 and FA9550-15-1-0254.

6. References

1. P. G. Soukiassian, "Wide band gap semiconductor: present status, future prospects and frontiers", *J. Phys. D: Appl. Phys.*, **40**, 20, 2007, p. 6138, doi: 10.1088/0022-3727/40/20/E01
2. S. Ghimire, G. Ndabashimiye, et. al. "Strong-field and attosecond physics in solids", *J. Phys B: At. Mol. Opt. Phys.*, **47**, 20, October 2014, 204030, doi: 10.1088/0953-4075/47/20/204030
3. B. H. Christensen, P. Balling, "Modeling ultrashort-pulse laser ablation of dielectric materials", *Phys. Rev. B*, **79**, April 2009, 155424, doi: 10.1103/PhysRevB.79.155424.
4. P. Balling, J. Schou, "Femtosecond-laser ablation dynamics of dielectrics: basics and applications for thin films", *Rep. Prog. Phys.*, **76**, 3, March 2013, 036502, doi: 10.1088/0034-4885/76/3/036502
5. M. Sun, J. Zhu, Z. Lin, "Modeling of ablation threshold dependence on pulse duration for dielectrics with ultrashort pulsed laser", *Opt. Eng.*, **56**, 1, January 2017, 011026, doi: 10.1117/1.OE.56.1.011026
6. L. V. Keldysh, "Ionization in the field of a strong electromagnetic wave", *Sov. Phys. – JETP*, **20**, 5, May 1965, pp. 1307-1314.
7. V. Vinogradov, "Absorption of high-intensity light by free carriers in dielectrics", *Sov. Phys. – JETP*, **41**, 3, 1975, pp. 540-543.
8. A. V. Vinogradov, "On the Drude formula for semiconductors", *Sov. Phys. – JETP*, **43**, 3, 1976, pp. 521-526.
9. D. Austin, K. Kafka, C. Blaga, L.F. Dimauro, and E. A. Chowdhury, "Measurement of femtosecond laser damage thresholds at mid IR wavelengths", *Proc. SPIE*, **9237**, October 2014, 92370V, doi: 10.1117/12.2068205.
10. S. Adachi, *Properties of Group IV, III-V, and II-VI Semiconductors*, John Wiley & Sons, Ltd., Chichester, 2005, doi: 10.1002/0470090340
11. L. A. Emmert, W. Rudolph, "Femtosecond laser induced damage in dielectric materials", Ch. 5 in *Laser-Induced Damage of Optical Materials*, D. Ristau, Ed., CRC Press/Taylor and Francis Group LLC, New York, December 2014, ISBN: 978-1-4398-7216-1.
12. N. W. Ashcroft, N. D. Mermin, *Solid State Physics*, Saunders College Publishing, New York, 1976, ISBN: 9780030493461
13. V. Gruzdev, "Analysis of the transparent-crystal ionization model developed by L. V. Keldysh", *J. Opt. Technol.*, **71**, 8, August 2004, pp. 504-508, doi: 10.1364/JOT.71.000504
14. V. Gruzdev, "Photoionization rate in wide band-gap crystals", *Phys. Rev. B*, **75**, 20, May 2007, 205106, doi: 10.1103/PhysRevB.75.205106