Ultrafast electron emission and associated electron dynamics from low dimensional nanostructures

Ph.D. Thesis

Author:
Madas Saibabu

Supervisor:
Mousumi Upadhyay Kahaly (Ph. D.)
ELI-ALPS, ELI-HU Non-Profit Ltd., Wolfgang Sandner utca 3., Szeged, H-6728, Hungary

Co-supervisor:
Subhendu Kahaly (Ph. D.)
ELI-ALPS, ELI-HU Non-Profit Ltd., Wolfgang Sandner utca 3., Szeged, H-6728, Hungary

Doctoral School of Physics
Institute of Physics
University of Szeged
Faculty of Science and Informatics
2023
Szeged

Introduction

Materials manifest a diverse array of electronic phases, encompassing categories such as metals, semimetals, semiconductors, superconductors, and insulators [1–4]. Of particular intrigue is the observation that the physical attributes of these systems can be substantially modulated by external parameters. The investigation of a material's response within a laboratory environment, specifically its response to external perturbation such as temperature fluctuations, structural modification, chemical modification, interfacing with other material, external field, etc is of paramount significance in the comprehensive assessment of the material's stability, functionality, and its potential applications. Electrons, being light subatomic particles with a charge, respond rapidly to temperature variations. In materials, electronic motion represents an ultrafast phenomenon, occurring on timescales in the order of sub-femtoseconds to attoseconds ($10^{-15} - 10^{-18}$ seconds) [5], thanks to their low mass and high mobility. In contrast, ion motion is considerably slower, taking place on the timescale of picoseconds (10^{-12} seconds) or longer due to their comparatively heavier mass, demonstrating a stark difference in temporal dynamics. Consequently, understanding and manipulation of ultrafast electronic processes in materials can greatly influence the materials response, like electron emission phenomena and external field-induced effects.

The ultrafast electron emission and electron dynamics depends on many factors, such as the nature of the material, i.e. the electronic structure, size, dimensionality, the type of external perturbation, among many others. Advances in electron emission materials and physics are driving a renaissance in science and technology, opening up new applications, such as energy conversion [6, 7] and ultrafast electronics [8], as well as improving traditional applications in electron imaging and high-energy science. These requirements have propelled the study of new materials and their emission mechanisms, often taking advantage of the unique electronic and thermal properties of low dimensional materials and nanoscale phenomena [8–10]. In contrast with their bulk counterparts, low dimensional structures may provide near-instantaneous electron emission [7, 11, 12]. They are fabricated from their bulk counterparts, resulting in spatial confinement [13] of the motion of electrons in a crystal in one, two or even all three directions. With adequate confinement, quantum mechanical effects lead to the discretization of the energy spectrum, i.e. the quantization of allowed energy levels and the momenta of electrons. Such quantization has profound effects on the ground state properties of these systems, as well as on the coupling between the electronic, nuclear and spin degrees of freedom [14], which consequently dictates the dynamic behavior too. The dimensionality of a material plays a crucial role in determining its electronic properties and ultrafast electron dynamics. Understanding and controlling these dynamics enable the design of novel materials and devices with enhanced functionalities for a wide range of technological applications.

Since the groundbreaking discovery of graphene in 2004 [15], the world of two-dimensional (2D) materials have been continuously expanding, comprising a vast range of condensed matter phases, such as metals [1], semiconductors [2, 16], insulators [17], semimetal [15], topological semimetal [3, 18], superconductor [19], Mott insulator [20], Bose-Einstein condensates [21]. 2D materials with atomic-thin thickness have gained immense popularity as a revolutionary material class that hold considerable potential in solid state technology. Recent advancements in fabrication techniques and an enhanced understanding of the fundamental physical properties of 2D materials have paved the way for the successful integration of 2D materials based components across a broad range of device applications. These applications span a wide spectrum, encompassing fields such as optoelectronics

[22], nanoelectronics [22], plasmonics [23], photodetection [24], nonlinear optics [25, 26], nanophotonics [27, 28], vacuum electronics [11], energy storage and conversion [29–31], and extending to emerging device paradigms like valleytronics [32], spintronics [33], twistronics [34], artificial neurons [35], and quantum emitters [36]. One of the significant features of 2D materials that are absent in their bulk counterparts is the possibility of mixing and matching 2D materials to obtain heterostructure with desirable properties.

One of the key factors which influences the ultrafast electron dynamics in materials is the external perturbation, as it introduces a powerful means to control and explore the behavior of electrons within materials [37]. External perturbations are of different kinds, including time-dependent laser fields (AC field), monochromatic field (DC field), temperature gradients, among many others. Ultrafast perturbations, such as femtosecond laser pulses (AC field), enable the investigation of ultrafast electron dynamics, offering insights into processes occurring on incredibly short timescales. This is pivotal for understanding energy relaxation and carrier dynamics in materials. Among several possible scenarios, the excited electrons can either escape in to the vacuum [38] or recombine with the parent ion, resulting in various ultrafast processes, such as high harmonic generation [39], etc. Furthermore, monochromatic pulses (DC field) play a crucial role in electron emission studies. A monochromatic pulse refers to electromagnetic radiation, often in the form of light, where all the photons have the same energy or frequency. The use of monochromatic pulses in electron emission studies provides a versatile and powerful tool to investigate electronic properties, excite specific states, and manipulate quantum behavior. Their precise energy control and selective excitation capabilities contribute to a deeper understanding of electron emission mechanisms and enable the exploration of material properties at the atomic and subatomic levels. To have an in-depth understanding on the behaviour of the materials under the influence of AC and DC fields, an integrated study is essential by considering suitable theoretical approaches and simulations, which is addressed in this study.

In my thesis, the primary aim is to understand the possible ways to tune and engineer the electron emission processes, by investigating diverse electron emission phenomena in relation to different materials and dimensionalities. As obvious, the electron emission process in a material is profoundly influenced by the internal electron dynamics within that material, such as energy distribution and thermalization of electrons, many-body effects that can alter the electronic band structure [6, 40] and affect the efficiency of electron emission mechanisms. Indeed many-body interactions, in particular exchange or correlation interactions between multiple electrons, play a significant role in the behavior of many atomic, molecular, and solid systems. The effects can manifest as large variations in binding energies, excitations, linear and nonlinear response and many other chemical/physical properties. Consequently, ultrafast electron dynamics within a material are affected.

Ultrafast electron dynamics captures the imprint of material's electronic and structural properties, providing valuable insights into how they respond to external stimuli and revealing the fundamental characteristics that define their behavior on extremely short timescales. This imprint encompasses information about energy transfer processes, carrier dynamics, and the interaction of electrons with the material's lattice structure, all of which are essential for understanding and manipulating the electronic properties of materials [7, 41]. Thus a combined understanding of electron emission and electron dynamics in materials have the power to influence not only the energy distribution but also the mechanisms governing electron ejection, leading to a spectrum of emission processes like photoemission, or thermionic emission. Thus, I also attempt to understand the ultrafast electron dynamics in atoms and low-dimensional solid materials, which is not only crucial for fundamental research but also has far-reaching implications in the development of advanced electron sources, photodetectors, and high-speed electronics, shaping the landscape of modern technology and scientific exploration.

It is important to note that conducting real-time experiments comes with inherent complexities, and access to large-scale experimental facilities which often are difficult to access. Through the utilization of numerical simulations and appropriate theoretical models, as attempted in my thesis, we can gain a comprehensive understanding of intricate details and provide complementary knowledge, valuable support to experimental observations. This dissertation, driven by simulations, delves into the intricate realm of electron dynamics within atoms, metals, and two-dimensional nanostructures when subjected

to external perturbations, including both DC and AC fields.

Methodology

In this section, I will briefly describe the semi-analytical and state-of-the-art quantum mechanical approaches, which I used to investigate the electron emission properties from metals, semiconductors, and ultrafast electron dynamics in 2D materials. First, I calculated the electron emission from coated metal surfaces at finite temperatures and explored the dependence of electron emission on factors, such as various metallic coatings, work function and thickness of the coating material, and the operating temperature. We developed a framework for determining and analyzing electron emission from a metal-coated target. This framework is based on Fowler's approach to electron emission, incorporates Fermi-Dirac statistics regarding the distribution of electron energy, utilizes a three-dimensional parabolic energy model for the base metal, and accounts for energy dispersion dependent on the thickness of the coating metal.

Following Fowler's approach to the statistical distribution for bulk metallic materials, the total number of electrons impinging the surface normally (for the parent surface) per unit area per unit time, with normal and parallel energy components in the range of (E_x and $E_x + dE_x$) and (E_t and $E_t + dE_t$), can be expressed as

$$d^2n_a = (A_0/e)T^2[T_a(e_x)F(e_x + e_t + \varphi_a - \upsilon_0 - \upsilon_{sc})]de_x de_t \ , \tag{1}$$

where $A_0 = 4\pi emk_B^2/h^3 \sim 117A/cm^2K^2$, $e_{x,t} = E_{x,t}/k_BT$, $\varphi_a = e\phi_a/k_BT$, $\upsilon_0 = eV_0/k_BT$, and (A_0/e) represents the flux associated with the Richardson constant (A_0). The term $\upsilon_{sc} = (e^3\mu)^{1/2}/k_BT$ accounts for the lowering of the potential barrier due to the Schottky effect, which causes a rise in particle energy. The parameter $\mu = 2V_0/d$ represents the electric field strength. The function $F(E)$ corresponds to the electron energy distribution, and T represents the surface temperature. The Boltzmann constant is denoted as k_B. After applying the appropriate particle distribution, such as Fermi-Dirac (FD), the Equation 1 can be simplified by integrating over the e_t space, which ranges from 0 to infinity. The resulting expression is given as

$$dn_{a,FD} = (A_0/e)T^2[T_a(e_x)\ln\left[1 + \exp\left(\upsilon_0 + \upsilon_{sc} - \varphi_a - e_x\right)\right]]de_x \ , \tag{2}$$

Equation 2 is used to calculate the flux of electrons that can surpass the triangular barrier present at the interface region. To calculate the electron population density within the coating material, we employ a non-parabolic energy dispersion relation that is dependent on the thickness, similar to that of 2D graphitic heterostructures [42]. The parallel dispersion relation for the top layer of the coating, with thickness (s), can be described as [42]

$$k_t dk_t = \left(\frac{\pi\hbar}{k_B}\right)(2A_0T/e)\left(\frac{e_h^{-2a_0/s}}{s/a_0}\right)(E - E_x)^{(2a_0/s)-1}dE. \tag{3}$$

The total energy of electrons inside the layer is given by $E = (E_x + E_t)$. The hopping parameter between consecutive atomic layers is denoted by e_h, while a_0 represents the interatomic mean distance. By using the above dispersion relation 3, the number of electrons hitting the top coated layer from inside with a total energy between E and $(E + dE)$ and a normal energy between E_x and $(E_x + dE_x)$ per unit area per unit time can be expressed as [7, 43, 44]

$$d^2n_b = (2A_0/e)T^2\left[e_h^{'-2a_0/s}/(s/a_0)\right](e - e_x)^{(2a_0/s)-1}F(e + \varphi_b + \upsilon_0)de\ de_x \ , \tag{4}$$

By integrating the distribution over the total energy range (e_x, ∞) and simplifying it further, the electron distribution with *FD* statistics for normal energy can be expressed as

$$dn_{b,FD} = (2A_0/e)T^2 \left[\frac{e_h^{'-2a_0/s}}{(s/a_0)}\right] \Gamma(2a_0/s) \left[\text{-Polylog}[2a_0/s, -\exp\left[-(e_x + \varphi_b + \upsilon_0)\right]]\right] de_x \ , \tag{5}$$

The total normal electron flux available for emission from the top layer with thickness s can be determined by combining the contributions from both the inner parent metal as given in Equation 2 and the outer surface layer population as given in Equation 5, and can be written as

$$dn_{FD} = dn_{a,FD} + dn_{b,FD} \ , \tag{6}$$

To obtain the total electron flux emitted from the coated surface, we need to integrate the normal flux obtained from the tunneling probability of the outer region's enhanced step barrier over the appropriate normal energy space, that is $e_x \equiv (0, \infty)$ as

$$n_{FD} = \int_0^\infty T_b(e_x)(dn_{a,FD} + dn_{b,FD})de_x \ . \tag{7}$$

Equation 7 has been numerically analyzed for a parametric study. The analysis suggests that the electron emission flux can be fine-tuned by selecting appropriate physical parameters of the coating substrates and base material. Using this approach I reported the results from our publication [T1] and summarized as Thesis point 1.

Next, I investigated the thermionic flux from a promising two-dimensional nanostructure, which was recently synthesized [45], for which we have combined an *ab-initio* quantum simulation tool, i.e. density functional theory (DFT) and semiclassical description of emission. DFT is an approach to transform the complex many-body Schrödinger equation into a set of single particle equations known as Kohn-Sham equations, which are easier to solve. DFT can make the exact transformation from the many-body Schrödinger equation to Kohn-Sham equations without any loss of information, provided one knows a mathematical object, called exchange-correlation function. DFT demonstrates that such a mathematical object exists but does not represent what it looks like. The main idea behind DFT is to use the particle density $n(r)$ as the key variable to calculate all the other observables. In other words, the knowledge of ground-state particle density implies knowledge of the wave function and the potential, and consequently, one can calculate all the observables. DFT is based on two mathematical theorems developed by Hohenberg and Kohn [46]:
Theorem I: For a system of interacting particles in the presence of external potential $V_{ext}(r)$, the potential $V_{ext}(r)$ is determined uniquely, except for a constant, by the ground-state particle density $n_o(r)$.
Theorem II: For any external potential $V_{ext}(r)$, a universal functional for energy $E[n(r)]$ can be defined. The density $n(r)$ that minimizes this functional is the exact ground-state density $n_o(r)$. According to the above two theorems, there is a one to one mapping between ground-state particle density $n_o(r)$ and external potential V_{ext}, and vice versa. With the knowledge of $n_o(r)$, one can obtain V_{ext}, with which we can construct the Hamiltonian, which is used to determine the wave function; thus all the properties of the system can be determined based on the ground-state density.

Based on density functional theory calculations using quantum espresso package [47], we determine the electronic bands and the band edges in phosphorene, followed by establishing a suitable dispersion relation using the tight-binding model. By using Fowler's approach for electron emission and incorporating the Fermi-Dirac statistics for electrons, the expression to calculate thermionic emission and photo-thermionic flux are derived.

The incident radiation flux on the surface of phosphorene has a dual effect: a portion of the absorbed energy causes surface heating through collision, while the remaining photon flux induces direct electron photoemission. The electrons in the valence and conduction bands of phosphorene

follow the Fermi-Dirac distribution. The surface can be biased by applying an external potential or due to continuous electron emission and recollection equilibrium. This bias is represented by potential V_s, which tunes the effective barrier height for electron emission as $V_T = \phi + V_s$. Under the influence of a finite positive potential, the energy structure drops down by energy $-eV_s$. We can express the distribution of momentum of the electron flux that approaches the top layer surface of phosphorene perpendicularly (at $z = 0$) from the inside and is available for emission, with a total energy between E_t and $(E_t + dE_t)$ and a normal energy (along the $\hat{z}$ axis, normal to the surface) between E_z and $(E_z + dE_z)$ as

$$d^3n = \left(\frac{\hbar k_z}{m}\right)\left(\frac{1}{2\pi^2}\right)F(E)dk_xdk_ydk_z = \left(\frac{k_BT}{2\pi^2\hbar a_0^2}\right)F(E)dk_xdk_ydk_z \ , \tag{8}$$

where $F = F_{FD} = [1 + \exp[\epsilon_z + \epsilon_t - \epsilon_f]]$ refers to FD distribution of electrons, $k = ka_0$, $\epsilon_t = E_t/k_BT$, $\epsilon_z = E_z/k_BT = \hbar^2k_z^2/2mk_BT$, $\epsilon_f = E_f/k_BT$, E_f refers to the Fermi energy level, a_0 is the interatomic distance between consecutive layers, $\hbar$ and k_B are the reduced Planck's and Boltzmann's constants, respectively, and T is the temperature of the electron emitting surface. The substitution of $(\hbar k_z/m)dk_z$ by $\hbar^{-1}dE_z$ has been made using the group velocity relation $\hbar k_z/m = dE_z/dk_z$ in order to obtain Equation 8. Upon simplifications that are explained in Chapter 4, to estimate the thermionic emission, the momentum distribution of the electrons within the sheet can be represented as follows:

$$d^3n_{Th} = f_T\left(\frac{k_BT}{2\pi^2\hbar a_0^2}\right)\left[1 + \exp(\epsilon_z - \epsilon_f)\exp\left[\Gamma^4(\mathbf{k}) + Abs[\Gamma^0(\mathbf{k})]\right]\right]^{-1}dk_xdk_ydk_z \ , \tag{9}$$

where n_{Th} is the thermionic emission flux. The momentum distribution of the electrons, which have absorbed a photon and have normal energy $\epsilon_z' = \epsilon_z + \epsilon_\nu$ and transverse momenta $k_x' = k_x$, $k_y' = k_y$ can be obtained by expressing ϵ_z, k_x, k_y in Equation 9 in terms of ϵ_z', k_x', k_y'. The resulting distribution can be written as

$$d^3n_{Ph} = f_P\left(\frac{k_BT}{2\pi^2\hbar a_0^2}\right)\left[1 + \exp(\epsilon_z - \epsilon_f - \epsilon_\nu)\exp\left[\Gamma^4(\mathbf{k}) + Abs[\Gamma^0(\mathbf{k})]\right]\right]^{-1}dk_xdk_ydk_z \ , \tag{10}$$

where n_{Ph} is the photo-thermionic emission flux. To obtain the flux resulting from the thermionic/photo-thermionic emission, we need to integrate the expressions (Equation 9, and Equation 10) over appropriate boundaries in **k**-space and the effective surface potential barrier (ϵ_z). The resulting net thermionic/photo-thermionic flux can be written as:

$$n_{Th,Ph} = \int d^3n_{Th,Ph} = \int_{\epsilon_c - \upsilon_s}^{\infty}\left(\int_{-k_{x0}}^{k_{x0}}\int_{-k_{y0}}^{k_{y0}} d^3n_{Th,Ph}dk_xdk_y\right)d\epsilon_z. \tag{11}$$

With the numerical simulations, I have prescribed how the electron emission flux can be further enhanced by photon irradiation and validated with the experimental findings. I have reported these results from our publication [T2] and summarized in Thesis point 2.

Next, I use a nonequilibrium model, i.e. two-temperature model (TTM) to describe the temporal evolution of electronic temperature, lattice temperature, electron thermalization duration, and electron-lattice thermalization duration along with the electron emission process from a metallic surface driven by an ultrafast laser pulse. First, we employ an enhanced 3D TTM that incorporates a nonlinear heat equation and laser interactions in all the three dimensions (*x*,*y*,*z*).

The 3D TTM is applied to model the spatio-temporal evolution of temperature and energy exchange among the electrons $T_e(x, y, z, \Delta t)$, lattice $T_l(x, y, z, \Delta t)$ of the solid, and the substrate $T_s(x, y, z, \Delta t)$, driven by a single Gaussian femtosecond laser source $Q(x, y, z, t)$, by considering the following 3D non-linear coupled parabolic equations, which are given as [48]

$$C_e(T_e)\frac{\partial T_e(x,y,z,t)}{\partial t} = \nabla.\big(k_e(T_e,T_l)\nabla T_e(x,y,z,t)\big) - G\big(T_e(x,y,z,t) - T_l(x,y,z,t)\big) + Q(x,y,z,t) - S_{es}(T_e,T_s), \tag{12a}$$

$$C_l\frac{\partial T_l(x,y,z,t)}{\partial t} = \nabla.\big(k_l\nabla T_e(x,y,z,t)\big) + G\big(T_e(x,y,z,t) - T_l(x,y,z,t)\big) - S_{ls}(T_l,T_s), \tag{12b}$$

$$C_s\frac{\partial T_s(x,y,z,t)}{\partial t} = \nabla.\big(k_s\nabla T_s(x,y,z,t)\big) + S_{ls}(T_l,T_s) + S_{es}(T_e,T_s), \tag{12c}$$

where C_e/C_l (C_s) and k_e/k_l (k_s) denote specific heat capacity and thermal conductivity of the electrons/lattice (substrate), respectively. G is electron-lattice coupling factor. $S_{es/ls}$ is the boundary interface heat exchange between solid electrons/lattice and the substrate, given as $S_{es/ls} = G_{es/ls}(T_{e/l} - T_s)$, where $G_{es/ls}$ is the corresponding thermal boundary conductance. Equation 12a describes the energy absorption of the electron subsystem from the laser pulse, the heat diffusion among the electrons, and heat transfer to the lattice. The Equation 12b is for the lattice subsystem, and contains a heat diffusion term, and energy input term due to coupling with the electrons. Equation 12c is for the substrate system which contains the diffusion term and the boundary interface heat transfer between the metal electrons/lattice and the glass substrate. The laser power density is denoted by $Q(x,y,z,t)$, which is the source of energy input into the electron subsystem and is considered as a Gaussian pulse, in time and space [49], directed at an angle θ (w.r.t normal of the sample) onto the 3D volume surface.

The electric field $E(x,y,t)$ is obtained by transforming the incident laser field to the sample coordinate system (x,y) and thus the subsequent distribution of intensity on the surface of the target is attained and given as $I(x,y,t) = \epsilon_0 c/2|E(x,y,t)|^2$, where ϵ_0 represents the permittivity of vacuum and c denotes the speed of light in vacuum. Nonetheless, when the laser is incident at an oblique angle, to conserve the energy that is deposited and spread out over an area larger than that of normal incidence, the intensity is scaled by a factor of $1/\cos\theta$. But, in this work, since we are conserving the intensity, $1/\cos\theta$ factor is not necessary to include. Thus, the effective intensity 'I_{eff}' can be written as $I_{\text{eff}}(x,y,t) = \epsilon_0 c/2\ |E(x,y,t)|^2$. Additionally, it is also necessary to include a suitable model for laser absorption at the areas near to the metal surface, taking into account the dynamic reflectivity $R_{s,p}(x,y,t)$, where s and p denotes the polarization of light that is perpendicular and parallel to the plane (x,z), respectively. The decay of Q as a function of z follows an exponential trend, in accordance with the physical principles that regulate the penetration of the pulse into the target.

$$Q\,(x,y,z,t) = \frac{I_{abs}(x,y,t)}{(\delta+\delta_b)(1-\exp\,(-d/(\delta+\delta_b)))}\exp\left(-\frac{z}{(\delta+\delta_b)}\right), \tag{13}$$

here I_{abs} represents the laser intensity absorbed by the sample at a point (x,y), which is given by $I_{\text{abs}}(x,y,t) = (1 - R_{s,p}(x,y,t))I_{\text{eff}}(x,y,t)$. Thickness of the target material is denoted with d, δ is the laser penetration depth, which depends on the wavelength and electron temperature, and is given as $\delta = 1/\alpha$ [50], where $\alpha = (4\pi Im(n_2))/\lambda$ is the absorption coefficient, imaginary part of the refractive index (n_2) is denoted by $Im(n_2)$, explained later. λ denotes the laser wavelength. To consider the effects of both diffusion and ballistic motion of hot electrons, the electrons' ballistic range, δ_b is considered to be 105 nm [51], and is included along with the optical penetration depth δ. As discussed earlier, during the laser-target interaction, absorbed intensity (I_{abs}) in Equation 13 is computed from

the reflectivity, which is modeled using Fresnel equations for S (R_s), and P-polarized light (R_p) [52]:

$$R_{\rm s}(x,y,t) = \left| \frac{n_1\cos\theta - n_2\sqrt{1-\left(\frac{n_1}{n_2}\sin\theta\right)^2}}{n_1\cos\theta + n_2\sqrt{1-\left(\frac{n_1}{n_2}\sin\theta\right)^2}} \right|^2 , \tag{14a}$$

$$R_{\rm p}(x,y,t) = \left| \frac{n_1\sqrt{1-\left(\frac{n_1}{n_2}\sin\theta\right)^2} - n_2\cos\theta}{n_1\sqrt{1-\left(\frac{n_1}{n_2}\sin\theta\right)^2} + n_2\cos\theta} \right|^2 , \tag{14b}$$

here refractive index of the ambient medium is denoted by n_1, considered to be unity (because it corresponds to vacuum), sample's refractive index $n_2 = n + ik = \sqrt{\epsilon_{DCP}(\omega)}$), where the real and imaginary parts of the refractive index are denoted by n and k, respectively. Gold's complex dielectric constant is denoted by ϵ_{DCP}. To calculate the dielectric constant, we used temperature-dependent electron relaxation times from the work of Block et al. [48] and combined it with the Drude-critical points (DCP) model. The model considered by Vial combines the Drude intraband contributions to permittivity with two-critical points model that account for interband transitions, given as [53]

$$\epsilon_{\rm DCP}(\omega) = \epsilon_\infty - \frac{\omega_P^2}{\omega^2 + i\gamma\omega} + \sum_{p=1}^{2} A_p\Omega_p \left(\frac{e^{i\phi_p}}{\Omega_p - \omega - i\Gamma_p} + \frac{e^{-i\phi_p}}{\Omega_p + \omega + i\Gamma_p} \right) . \tag{15}$$

In Equation 15, the right-hand side consists of three terms, each with specific contributions to the model. The first two terms represent the standard Drude model [54], where ϵ_∞ is the high-frequency limit dielectric constant, the damping term is denoted by γ and the plasma frequency $\omega_P = \sqrt{\frac{e^2 n_e(T_0)}{\varepsilon_0 m_{eff}} \frac{1}{(1+\beta\Delta T_l)}}$, where e is the charge of an electron, the ambient temperature is denoted by T_0, free electron density is denoted by $n_e(T_0)$ that is equivalent to $5.9 \times 10^{28}\ m^{-3}$, ε_0 is the vacuum dielectric permittivity, m_{eff} is the effective mass of an electron ($\sim 1.094 \times m_e$, the mass of an electron [55]), and β is the volume thermal expansion coefficient of gold, which is approximately $4.23 \times 10^{-5}\ K^{-1}$ [56]. The third term in Equation 15 accounts for two interband transitions and employs the two-critical points transition model as explained in the reference [57]. ϕ_p denotes the phase, Ω_p signifies the energy of the gap, $A_p\Omega_p$ represents the dimensionless amplitude, and Γ_p represents the broadening, which is given as $\Gamma_p = A_{\Gamma p}T_e^2 + B_{\Gamma p}T_l + \gamma_p$. The electron collision rate term γ can be separated into two components: electron-electron collision rate γ_{e-e} and electron-phonon collision rate γ_{e-ph}, where the overall rate is given by $\gamma = \gamma_{e-ph} + \gamma_{e-e}$, with $\gamma_{e-e} = AT_e^2$ and $\gamma_{e-ph} = BT_l$ [48, 58]. The parameters for Equation 15 resulting from the fit are displayed in Table 1. Using this model, I presented the findings from our publication [T3] and summarized them as Thesis point 3.

Next, to study the laser-driven non-linear response in atoms and solids, I have used state-of-the-art *ab-initio* based time-dependent density functional theory (TDDFT) approach, which takes multielectrons effects in to consideration. In TDDFT, the time-dependent Schrödinger equation, known as the time-dependent Kohn-Sham (TDKS) equation is given as:

$$i\frac{\partial}{\partial t}\phi_j(\mathbf{r},t) = \Big[-\frac{\nabla^2}{2} + \upsilon_{KS}(\mathbf{r},t) \Big]\phi_j(\mathbf{r},t) \ . \tag{16}$$

The first term on the right hand side of Equation 16 is the kinetic energy operator.

$$\upsilon_{KS}(\mathbf{r},t) = \upsilon_{ext}(\mathbf{r},t) + \upsilon_H(\mathbf{r},t) + \upsilon_{\rm XC}(\mathbf{r},t), \tag{17}$$

$$= \upsilon_{ext}(\mathbf{r},t) + \int \frac{\rho(\mathbf{r}',t)}{|r-r'|} d\mathbf{r}' + \upsilon_{\rm XC}(\mathbf{r},t) \tag{18}$$

Parameter	Value
A $[K^{-2}s^{-1}]$	1.2 $\times 10^{7}$
B $[K^{-1}s^{-1}]$	4.428703071 $\times 10^{11}$
Ω_1 $[rad\ s^{-1}]$	4.01772608$\times 10^{15}$
$A_{\Gamma 1}$ $[K^{-2}s^{-1}]$	1.2 $\times 10^{7}$
$B_{\Gamma 1}$ $[K^{-1}s^{-1}]$	1.14681587030 $\times 10^{11}$
γ_1 $[rad\ s^{-1}]$	7.9 $\times$ 10^{14}
ϕ_1	$-\pi/4$ [55, 59]
$A_{\Gamma 2}$ $[K^{-2}s^{-1}]$	1.2 $\times 10^{7}$
$B_{\Gamma 2}$ $[K^{-1}s^{-1}]$	6.094240955631399 $\times 10^{11}$
γ_2 $[rad\ s^{-1}]$	1.9 $\times$ 10^{15}
ϕ_2	$-\pi/4$ [55, 59]
Ω_2 $[rad\ s^{-1}]$	5.56883141$\times 10^{15}$
A_1	0.917783355
A_2	1.52288304
ϵ_∞	1.197

Table 1: Parameters in Equation 15 calculated by fitting to Johnson and Christy data [60].

$\upsilon_{ext}(\mathbf{r},t)$ is the external potential including the external laser field and the Coulomb interaction between the electron and the nuclei, which is defined in the pseudopotentials. υ_H is the Hartree electrostatic interaction between the electrons. υ_{XC} is the time-dependent exchange-correlation potential including all the non-trivial many-body effects. The density of the interacting system is obtained from the orbitals ϕ_j by

$$\rho(\mathbf{r},t) = \sum_{j}^{N} |\phi_j(\mathbf{r},t)|^2 \ , \tag{19}$$

where the summation is over all the occupied states. A practical method to solve TDDFT equations is as follows. As an initial step to our calculation, we can select the TDKS Equation 16 and directly integrate them. Starting from the ground-state at time t = 0, one can calculate the density at time $t = t'$, $n(\mathbf{r},t)$ by propagating the occupied ground-state KS orbitals.

$$\phi_\upsilon(\mathbf{r},t') = \hat{U}(t',0)\phi_\upsilon(\mathbf{r}) \ , \tag{20}$$

where $\hat{U}$ is the time evolution operator

$$\hat{U}(t',0) = \hat{\mathcal{T}} \exp\left[-i \int_0^{t'} \hat{H}(\tau) d\tau \right] \ , \tag{21}$$

where $\hat{\mathcal{T}}$ is the time ordering operator. Using this approach, the density at time $t = t'$ can be written as $n(\mathbf{r},t') = \sum_\upsilon |\phi_\upsilon(\mathbf{r},t')|^2$. In practice, propagation is performed by splitting the time interval between $t = 0$ to $t = t'$ in to small steps δt using the property $\hat{U}(t_2,t_1) = \hat{U}(t_2,t_3)\hat{U}(t_3,t_1)$:

$$\phi_\upsilon(\mathbf{r},t+\delta t) = \hat{U}(t+\delta t,t)\phi_\upsilon(\mathbf{r},t) \ . \tag{22}$$

It is important to choose appropriate approximations for the time evolution operator $\hat{U}(t+\delta t,t)$ in real calculations, both in terms of numerical accuracy and computational cost. No matter which approximation is used, they should all accurately reproduce the exact properties of $\hat{U}$, such as unitary, which is important to ensure the convergence of the total charge during the propagation. The simplest approach is the midpoint rule, in which the propagator is approximated by the exponential of the Hamiltonian calculated at time $t + \delta t/2$, as

$$\hat{U}(t+\delta t,t) = \exp\left[-i\hat{H}(t+\frac{\delta t}{2})\delta t \right] \ , \tag{23}$$

where $\hat{H}(t+\frac{\delta t}{2})$ is calculated self-consistently with the predictor corrector method [61]. In the above equation, the exponential, to be calculated, must be expanded in Taylor series to a given order. Hence, Equation 22 can be written as

$$\phi_v(\mathbf{r}, t+\delta t) = \Big[1 - i\hat{H}(t+\frac{\delta t}{2})\delta t - \frac{1}{2}\hat{H}^2(t+\frac{\delta t}{2})\delta t^2 + ...\Big]\phi_v(\mathbf{r}, t) \ . \tag{24}$$

The high harmonic spectrum $I(\omega)$ for atoms is calculated from the Fourier transform ($\mathcal{FT}$) of the time-dependent dipole acceleration $\boldsymbol{a}(t)$ as [62–64]

$$I(\omega) = \left|\mathcal{FT}[\boldsymbol{a}(t)]\right|^2 = \left|\mathcal{FT}[\ddot{\boldsymbol{d}}(t)]\right|^2 \tag{25}$$

where $\boldsymbol{d}(t) = \int n(r,t) r dr$ is the time-dependent dipole moment, $n(r,t) = \sum_{i=1}^{N} |\phi_i(r,t)|^2$ is the time-dependent electron density and N is the number of Kohn-Sham orbitals given by $\phi_i(r,t)$. Using this approach I reported the findings related to non-linear response from bi-chromatic circular counter-rotating pulse-driven argon atom from our publication [T4] and summarized in Thesis point 4.

Furthermore, in case of high harmonic spectrum $I(\omega)$ in solids, we obtain total electronic current in the system, $\boldsymbol{J}(t) = \partial/\partial t \int_\Omega d^3r \boldsymbol{j}(r,t)$, which is obtained by integrating the microscopic electronic current density, $\boldsymbol{j}(r,t)$, over the unit cell, where Ω is the unit cell volume [65]. The microscopic electron current can be written as

$$\partial/\partial t \int_\Omega d^3r \boldsymbol{j}(r,t) = -\int_\Omega d^3r n(r,t) \nabla v(r,t) \ , \tag{26}$$

where $n(\mathbf{r},t)$ is the time-dependent electron density of the material driven by laser field. External potential $v(r,t)$ corresponds to both the electron-nuclei potential $v_0(r)$ and the applied laser field. The HHG spectrum $[I(\omega)]$ is obtained by applying a discrete Fourier-transform ($\mathcal{FT}$) to $\boldsymbol{J}(t)$, which is given as

$$I(\omega) = \Big|\mathcal{FT}\big(\boldsymbol{J}(t)\big)\Big|^2 \ . \tag{27}$$

Using this method, I investigated the strong-field nonlinear optical properties of a freestanding 2D pristine phosphorene and strain induced band gap modified phosphorene systems. I presented our results from the article published in *arXiv* [T5] and summarized in the Thesis point 5.

Summary

Temperature dependent response of a material is crucial in assessing its stability, functionality and potential applications. With increase in the operating temperature in laboratory environment, the electronic properties and lattice vibrations in any material will modify. For instance, at 300 K or beyond, material surface separates the heated electrons within the material from otherwise non-conducting space. Such gradient promotes emission of electrons across the surface boundary and results in thermionic emission, and strongly depends on the work function, potential barrier, surface structure, electronic properties of the material. When transitioning from a bulk phase to a gas-like state, surface structure changes and the confinement effects [12, 66] can have significant implications on the behavior and properties of the material. Confinement effects arise when a material's dimensionality is reduced from bulk to two-dimensional or one-dimensional or zero-dimensional, altering its physical, chemical, and

thermodynamic characteristics. These effects are particularly pronounced when a material is confined to dimensions on the order of nanometers or smaller, leading to phenomena that differ from those observed in bulk systems. In nanoscale confinement, the quantization of energy levels becomes significant. This can lead to discrete energy states that affect electronic properties, influencing phenomena such as optical absorption and emission. Dimensionality of the material impacts the electronic structure of the ground state and consequently their time-dependent evolution under any external field.

As is well known, motion of electrons and ions are an ultrafast phenomenon ($\sim 10^{-15}$ s for electrons and $\sim 10^{-12}$ s for ions). Understanding and tuning ultrafast electronic processes in materials offer opportunities to unlock new scientific insights and technological breakthroughs with far-reaching impacts. These ultrafast processes can be captured with laser assisted advanced metrology techniques and show strong dependence on the intensity of the external perturbation. By probing different electron dynamics with respect to the different dimensionality will provide contrasting perspective. Any real-time experiment will have its own complexities. Numerical simulations and suitable theoretical models helps in understanding the intricate details and provides support to the experiments. In this dissertation, using simulations, I have investigated the electron dynamics in atoms, metals and two-dimensional nanostructures under the influence of an external perturbation, such as DC and AC field. In the following, I summarize the important findings in my doctoral thesis:

Thesis point 1

Electron emitters play a significant role in laser experiments, particularly in the field of ultrafast and high-intensity laser physics. In such experiments, intense electric fields results in laser-induced electron emission phenomena. These emitted electrons can be utilized in various applications, such as material characterization, particle acceleration, radiation sources, to name a few. Coatings play a crucial role in improving the efficiency of the electron emitters. To enhance the electron emission efficiency from metallic surfaces, I have investigated the dependence of electron emission on different physical factors, such as different metallic coatings, thickness and work function of the coating material, and the operating temperature. To estimate and analyze the electron emission from a coated metal target, we established a formalism based on Fowler's treatment of electron emission, along with Fermi-Dirac statistics of the electron energy distribution, a three-dimensional parabolic energy dispersion for the target metal and thickness dependent energy dispersion for the coating metal. The surface coating induces a high potential field resulting in enhanced electron emission flux. This formulation holds good for any metal/semiconductor combination. We particularly examined the role of thickness of the coating material which is found to play an important role on the emission properties. From our numerical simulations, I have determined that at higher temperatures (exceeding 1500 K), the electron emission flux becomes noticeably prominent for the thinner coating, whereas a thicker coating demonstrates enhanced emission efficiency at lower temperatures. Our results suggest that by tuning the thickness of the coating layer, it is possible to tune the electron emission operating at a desired operating temperature. The understanding of electron emission phenomena, and suitable estimation of current from the coated surfaces, as presented in this analysis, hold potential practical significance for the creation of effective and adaptable field emitters and thin film devices. These results were published in [T1]. However, confinement effects arises due to change in the dimensionality of the material. This demands a quantum description of low-dimensional material's characteristics, such as two-dimensional semiconductors, whose electron emission properties are probed in the next thesis point.

Thesis point 2

As mentioned earlier, two-dimensional materials offer several advantages compared to their bulk counterparts, such as tunable band gap, high carrier mobility, enhanced optical properties and mechanical strength, to name a few. To investigate the thermionic flux from a promising two-dimensional nanostructure, we choose a semiconductor that was recently synthesized [45], for which we

have combined an *ab-initio* quantum simulation tool and semiclassical description of emission. Based on density functional theory calculations, we determine the electronic bands and the band edges in phosphorene, followed by establishing a suitable dispersion relation using the tight-binding model. By using Fowler's approach for electron emission and incorporating the Fermi-Dirac statistics for electrons, the expression to calculate thermionic emission and photo-thermionic flux are derived. With the numerical simulations, I have demonstrated that black phosphorene, a two-dimensional allotrope of phosphorous, has potential to be an efficient photo-thermionic electron emitter. In addition, I have prescribed how the electron emission flux can be further enhanced by photon irradiation and validated with the experimental findings. Our simulation results suggest that due to the intrinsic anisotropic energy dispersion of phosphorene, it's emission flux is higher when compared to well established graphene. This makes two-dimensional phosphorene a viable contender for photo-thermionic emission and energy conversion technologies relying on thermionic emission. The present methodology offers a fundamental understanding of the photo-thermionic features of two-dimensional phosphorene, exhibiting features that matches well with the experimental findings. These results are reported in our published article [T2]. As mentioned earlier, the electron dynamics show strong dependence on the external perturbation. Often in experiments, a pulsed laser field is used to study the ultrafast electron dynamics in metals or semiconductors. The semi-analytical approach to describe material properties and external perturbation, i.e. monochromatic field must be appropriately modified to investigate the ultrafast laser driven electron dynamics in metals and semiconductors, which is further investigated and summarized in my next thesis point.

Thesis point 3

Efficient three-dimensional description of ultrafast laser pulse and its interaction with the material can lead to exact estimation of ultrafast thermal evolution processes. Femtosecond laser driven ultrafast thermionic emission results in electron bunches that have a wide range of applications, such as high-gain harmonic generation free electron laser, laser accelerators, among many others. I have investigated the thermal evolution in gold coated glass substrate irradiated by an ultrashort laser pulse. Gold being a noble [67] transition metal has wide range of applications ranging from biomedicine [68], twistronics [69], flexible integrated electronics [70]. Additionally, gold coated mirrors are present everywhere in the ultrafast laser optics. Ultrafast electron emission in metal nanofilms are important in studies that apply to all these applications. To determine the spatio-temporal evolution of electron and lattice temperatures, I have implemented a three-dimensional two-temperature model, which takes in to account the three-dimensional laser pulse profile focused obliquely on to the sample. The associated thermionic emission properties are determined using modified Richardson-Dushman equation with space-charge effects and are solved self-consistently in our approach. Furthermore, I have determined the role of laser polarization and its angle of incidence on the spatio-temporal evolution of electron and lattice temperatures, and subsequently on the thermionic electron emission processes. I showed the role of temperature dependent reflectivity on the laser energy absorption. The maximum surface electron temperature monotonically rises with laser incidence angle for P polarized laser, while an opposite trend is noticed in S polarized case. A strong dependence of thermionic emission duration on the laser incidence angle and contrasting polarization dependent behaviour is observed. Additionally, I showed the strong dependence of thermionic current duration on the intrinsic electron-lattice thermalization duration of the sample. The conclusions drawn from our analysis and insights help in understanding and tuning ultrafast thermionic emission and associated atomic-level mechanisms in metals and also in semiconductors that behave like metals upon interaction with ultrafast lasers. This understanding could lead to enhancement of both thermionic emission current and its duration, thereby contributing to the development of designing efficient ultrafast thermionic emitters. These results are reported in our published article [T3].

Thesis point 4

Laser-matter interactions not only affect the electron emission but also for each laser cycle, sub-cycle electron dynamics changes during the interaction period. These sub-cycle dynamics needs to be captured for better understanding of material's response, which could lead to high harmonic generation, which involves the non-linear interaction of the laser field with an atom, or a molecule, or a solid-state medium, that results in the harmonics emission that are integer multiples of the driving laser frequency. High harmonic generation has emerged as a promising technique because of its ability to generate high-frequency light in the extreme ultraviolet and soft X-ray regions of the electromagnetic spectrum. While the initial high harmonic generation models were often based on the single-active electron approximation or time-dependent Schrödinger equation approach, the inclusion of multielectron effects in the study of high harmonics generation in atoms is of paramount importance as it enables a more accurate and comprehensive understanding of the complex physical processes underlying this phenomenon. Hence, we use time-dependent density functional theory approach, which takes into account the multielectrons effects to investigate the ultrafast electron dynamics in Ar atom irradiated by a linear polarized laser and then a bi-circular counter rotating laser fields. I have validated our numerical approach with experimental findings [71], by obtaining a significant feature of the high harmonic generation spectrum of Ar atom, such as the presence of Cooper minimum at $\sim 48\ eV$.

Bi-chromatic circular counter rotating laser fields create an environment of extremely strong and complex electromagnetic interactions with matter. This makes them invaluable for exploring fundamental strong-field physics phenomena, such as high harmonic generation, non-linear optics, etc. The counter-rotating nature of these laser fields allows enhanced control over the electron trajectories and dynamics. By adjusting the relative phases and intensities of the two beams, we can steer and manipulate electronic processes with high precision. Through a combination of state-of-the-art experiments, semi-classical analysis, and time-dependent density functional theory simulations we have showcased an efficient way to generate and characterize highly energetic and elliptically polarized higher-order harmonics from argon [T4]. Using time-dependent density functional theory approach, I have shown that by appropriately tuning the central wavelength of the second harmonic, the central frequency of the high harmonic radiation is continuously tuned. Our numerical simulations support the experimental findings, which reveal that the highly elliptical HHG radiation from argon can be tuned for an energy range of $\Delta E \approx 150\ meV$ in the spectral range of $\approx 20\ eV$. We anticipate that the ability to adjust energetic and highly elliptical high harmonic generation spectra could eliminate the limitations imposed by the need for few-cycle driving pulses in the generation of isolated circular attosecond pulses. These results are reported in the article that was just accepted in the *Physical Review A* journal [T4].

Thesis point 5

Advancements in both fabrication techniques and the underlying fundamental knowledge of two-dimensional materials have paved the way for their effective utilization in a wide array of device applications ranging from nonlinear optics, optoelectronics, photodetection, and many more. As mentioned before, phosphorene is a relatively new, promising member in the family of two-dimensional materials has attracted immense attention in the research community due to its distinctive features such as an adjustable bandgap, high carrier mobility, and noteworthy intrinsic in-plane anisotropy. Now, using these inherent structural and electronic traits, we investigated the ultrafast electron dynamics and high harmonic generation from laser driven phosphorene surface. We explored the impact of external strain on the electronic band structure and associated nonlinear effects in phosphorene. Our analysis reveals that strong field-driven processes within such systems can be optimized and controlled through biaxial tensile (+ ve) and compressive strain (- ve) engineering, effectively modifying the electronic structure. The application of strain, ranging from -10% to 2%, yields intriguing outcomes. Specifically, a -10% strain induces bandgap closure, whereas a 2% strain increases the gap by 22%, relative to the pristine phosphorene value of $0.9\ eV$. These changes are found to significantly influence

the high harmonic generation output.

The closing of the bandgap due to strain, especially within the range of 2% to −10%, contributes to an increased electronic density of states near the Γ−point, which results in enhanced electronic excitation. The intrinsic in-plane anisotropy of phosphorene has proved to be a significant factor, with harmonic yield displaying higher intensity when the laser polarization aligns along the armchair direction compared to the zigzag direction across all strain conditions. Electronic band structure engineering through external application of compressive strain had resulted in an increased number of excited electrons, which eventually leads to an enhanced high harmonic generation. Particularly striking is the nearly three times more enhancement in harmonic yield achieved under −10% strain along the armchair direction. These results are uploaded in *arXiv* [T5] and will be soon submitted for publication in peer reviewed journal. This comprehensive study widens the horizons of phosphorene research, uncovering uncharted territory and highlighting its potential for applications in extreme-ultraviolet and attosecond nanophotonics, as well as an efficient table-top high harmonic generation sources.

In light of the key findings and the thesis points discussed above, the main significance of this doctoral thesis is to investigate and understand the effects, both in presence and absence of pulsed laser fields, on the electronic structure properties, electron emission, and the subsequent ultrafast electron dynamics in materials of different dimensions. Our results based on state-of-the-art simulation approaches and theoretical modeling are compared/understood with respect to recent experiments, particularly focusing on their utility in applied science and engineering. The thorough simulations-based investigation carried out in this thesis proposes ways to enhance the photo and laser-induced thermionic emission properties in metal and semiconductor nanostructures, which has wide range of applications, such as thermionic emission based high temperature solar converters, thermionic energy converters, high-brightness ultrafast electron imaging tools, to name a few. Using appropriate optimized computational tools and numerical methods presented in this dissertation, we investigated the photo-induced thermionic emission from metals and two-dimensional semiconductor nanomaterials and explored the potential of phosphorene and alike materials for applications as an stable table-top high harmonic generation sources. Often the results presented in my dissertation, for example the emission current estimation from our analytical model in the case of phosphorene is successfully tested against experiments. Additionally, high harmonic generation from argon under the influence of linear and bi-chromatic counter rotating laser fields were benchmarked with experimental findings, complimenting and explaining the experimental traits. With numerical simulations presented in this dissertation, we aim to mimic the real-time laser-matter experiments, bypassing experimental challenges that will provide a complementary tool to experiments to understand ultrafast electron dynamics in metals, atoms and two-dimensional materials for future applications. It is worth stressing that the theoretical approaches that were used in my dissertation are not limited to the materials reported here, but can also be extended to other similar metals/semiconductors. Therefore, we strongly believe that our results will encourage new experimental ventures to investigate ultrafast electron dynamics and enhance the subsequent electron emission properties in two-dimensional materials.

Magyar nyelvű összefoglaló

Egy anyag stabilitásának, funkcionalitásának és lehetséges alkalmazásainak felméréséhez elengedhetetlen az anyag fizikai hatásokra való hőmérsékletfüggő válaszának ismerete. Laboratóriumi körülmények között az anyag elektronikus tulajdonságai és rácsrezgései a hőmérséklet növelésével megváltoznak. Az anyag hőmérsékletének 300 K felé emelésével például az anyag felületén elkülönülnek az elek-

tronok az anyagon belül az egyébként nem vezető térben. Az így kialakuló gradiens elősegíti a felülethatáron lezajló elektronemissziót, így termionikus emisszióhoz vezetve. Ez erős függést mutat az anyag munkafüggvényétől, felületi potenciáljától, felületi szerkezetétől, és az anyag elektromos tulajdonságaitól. A szilárd fázisból gázszerű állapotba való átmenet során a felületi szerkezet változásai, valamint a kvantumbezárásos jelenségek jelentős kihatással bírhatnak az anyag viselkedésére és tulajdonságaira. Kvantumbezárásos jelenségek akkor lépnek fel, amikor az anyag dimenzionalitása háromdimenziósról kettő, egy, vagy nulla dimenziósra csökken, melynek hatására a fizikai, kémiai és termodinamikai tulajdonságai is megváltoznak. Ezek a hatások kimondottan erősen jelentkeznek akkor, ha az anyag kiterjedése nanométeres vagy az alatti méreteket ölt, olyan jelenségekhez vezetve, melyek különböznek a háromdimenziós anyagokban tapasztalhatóktól. Ez a kvantumbezárás diszkrét energiaállapotokhoz vezethet, befolyásolva az olyan jelenségeket, mint az optikai abszorpció és emisszió. Az anyag dimenzionalitása hatással van az alapállapot elektromos szerkezetére, és így annak egy külső tér hatása alatt lezajló időbeli fejlődésére is.

Mint az közismert, az elektronok és ionok mozgása ultragyors időskálán zajlik (kb. $\sim 10^{-15}$ s és $sim10^{-12}$ s karakterisztikus idővel). Ezeknek az anyagokban lezajló ultragyors elektromos folyamatoknak a megértésével lehetőséget kapunk új tudományos ismeretek megszerzéséhez, és sokrétű hatású technológiai áttöréseket érhetünk el. Az ilyen ultragyors folyamatok, melyek megfigyelése lézerrel támogatott korszerű mérési technikák segítségével kivitelezhető, erős függést mutatnak a külső hatás intenzitásától. A különböző elektrondinamikai folyamatok vizsgálata különböző anyagi dimenzionalitások esetében kontrasztos perspektívát nyújt. Minden különböző valós idejű kísérletnek sajátságos komplexitása van; azonban numerikus szimulációk és megfelelő elméleti modellek segítséget nyújtanak az összetett részletek megértésében, és segítséget nyújtanak a kísérletekhez. Ebben a doktori disszertációban szimulációk segítségével az elektronok dinamikáját vizsgáltam atomokban, fémekben és kétdimenziós nanoszerkezetekben külső perturbáció, például állandó és időfüggő elektromos tér hatására. A disszertáció egy rövid bevezetéssel kezdődik, mely az anyagokban lezajló ultragyors elektrondinamikai jelenségek jelentőségét, valamint az elektronemisszió dinamikáját és karakterisztikáját befolyásoló tényezőket mutatja be. Ezt a módszertani fejezet követi, melyben részletesen ismertetjük a disszertációban alkalmazott elméleti módszerek mindegyikét. Ezután kutatásom legfontosabb eredményeit fejtem ki a soron következő fejezetekben. Az alábbiakban doktori értekezésem főbb eredményeit foglalom össze:

1. tézispont

A lézerekkel folytatott kísérletekben, különösen az ultragyors és a nagyintenzitású lézerfizika területén, jelentős szerepet játszanak az elektronemissziós források. Az ilyen kísérletekben az intenzív elektromos terek lézerindukált elektronemissziós jelenségekhez vezetnek. A kibocsátott elektronok ezután többféle módon alkalmazhatók, például anyagelemzésre, részecskegyorsításra, vagy sugárforrásként. A bevonatok döntő szerepet játszanak az elektronemisszió hatékonyságának javításában. A fémfelületek elektronemissziós hatékonyságának növelése érdekében megvizsgáltam az elektronemisszió függését olyan különböző fizikai tényezőktől, mint a fémbevonatok anyagi minősége, a bevonóanyag vastagsága és munkafüggvénye, valamint a működési hőmérséklet. A bevonatos fém céltárgyakból származó elektronemisszió becslésére és elemzésére kidolgoztunk egy, az elektronemisszió Fowler-féle leírásán alapuló formalizmust, az elektronok energiaeloszlásának Fermi-Dirac-statisztikájának, a fém céltárgy háromdimenziós parabolikus energiadiszperziójának, és a bevonófém vastagságfüggő energiadiszperziójának figyelembe vételével. A felületi bevonat erős potenciálteret alakít ki, ami fokozott elektronemissziós fluxust eredményez. A kidolgozott formalizmus bármely fém/félvezető kombinációra érvényes. Munkánkban kiemelten vizsgáltuk a bevonóanyag vastagságának szerepét, amelyről bebizonyosodott, hogy fontos szerepet játszik az emissziós tulajdonságokban. Numerikus szimulációinkból megállapítotuk, hogy magasabb hőmérsékleten (1500 K felett) a vékonyabb bevonat esetében az elektronemissziós fluxus jelentősen megnő, míg a vastagabb bevonat alacsonyabb hőmérsékleten mutat fokozott emissziós hatékonyságot. Eredményeink arra utalnak, hogy a bevonatréteg vastagságának hangolásával a kívánt üzemi hőmérsékleten működő elektronemisszió hangolható. Az elektronemissziós jelenségek megértése,

valamint a bevonatolt felületekről származó elektronáram megfelelő becslése, ahogyan azt ebben az elemzésben bemutatjuk, potenciális gyakorlati jelentőséggel bír nagy hatékonyságú, adaptálható téremissziós források és vékonyréteg-eszközök létrehozásában. Ezeket az eredményeket, melyek publikálásra kerültek [T1], a soron következő pontban tárgyaljuk. A kvantumbezárásos jelenségek azonban az anyag dimenzionalitásának megváltozásából származnak. Ezek megkövetelik az olyan alacsony dimenziójú anyagok tulajdonságainak kvantumos leírását, mint a kétdimenziós félvezetők. Az ilyen anyagok elektronemissziós tulajdonságait a következő tézispontban tárgyaljuk.

2. tézispont

Ahogy korábban említettük, a kétdimenziós anyagok számos előnnyel rendelkeznek háromdimenziós társaikhoz képest. Ilyen előny többek között a tiltott sáv szélességének hangolhatósága, a magas töltéshordozó-mobilitás, a kedvezőbb optikai tulajdonságok, és a magasabb mechanikai szilárdság. Hogy egy ígéretes kétdimenziós nanoszerkezetből származó termionikus áramlást vizsgáljunk, egy, a közelmúltban szintetizált félvezetőt választottunk [45], amelynek leírásához kombináltunk egy *ab-initio* kvantumszimulációs módszert az emisszió félklasszikus leírásával. Sűrűségfunkcionál-elméleti számítások segítségével meghatároztuk az elektronsávokat és a sávszéleket a foszforénben, majd egy megfelelő diszperziós összefüggést állítottunk fel a szoros kötésű modell segítségével. Fowler módszerét alkalmazva az elektronemisszióra, valamint az elektronokra vonatkozó Fermi-Dirac statisztika figyelembe vételével levezettünk egy képletet a termionikus emisszió és a foto-termionikus fluxus meghatározására. Numerikus szimulációk segítségével megmutattam, hogy a fekete foszforén, a foszfor kétdimenziós allotrópja, potencionálisan hatékony foto-termionikus elektronemissziós forrás lehet. Emellett megmutattam, hogy az elektronemissziós fluxus tovább növelhető fotonbesugárzással, mely eredményt kísérleti eredmények igazolnak. Szimulációs eredményeink azt mutatják, hogy a foszforén eredendő anizotropikus energiájának köszönhetően emissziós fluxusa magasabb a már jól ismert grafénénál. Ez az eredmény a kétdimenziós foszforént esélyes versenyzővé teszi a foto-termionikus emisszió területén, és az ezt alkalmazó energiakonverziós technológiákban. Jelen módszertan alapvető megértéssel ruház fel bennünket a kétdimenziós foszforén foto-termionikus tulajdonságait illetően, kísérleti eredményeknek megfelelő jellemzőket mutatva. Ezen eredményeink publikálásra kerültek [T2]. Azonban, amint korábban említettem, az elektrondinamikát jelentősen befolyásolják a külső perturbációk. A kísérletek során gyakran lézerimpulzusokat alkalmaznak a fémes vagy félvezető anyagokban lezajló ultragyors elektrondinamika tanulmányozására. A korábbi pontokban bemutatott, az anyagi tulajdonságok és a külső perturbáció (monokromatikus tér) hatását leíró félanalitikus modellünket módosítanunk kell úgy, hogy az megfelelően leírja az ultragyors lézer által vezérelt elektrondinamikát fémekben és félvezetőkben. Ezt a soron következő fejezetekben tárgyaljuk.

3. tézispont

Az ultragyors háromdimenziós lézerimpulzusok, valamint anyaggal való kölcsönhatásuk hatékony háromdimenziós leírása lehetővé teszi az ultragyors hőmérsékletváltozás pontos becslését. A femtoszekundumos ultragyors termionikus emisszió által olyan elektroncsomagok keletkeznek, melyek a magas hatásfokú szabadelektron-lézerektől a lézergyorsítókig számos területen alkalmazhatók. Munkámban vizsgáltam a hőhatás időbeli fejlődését ultrarövid lézerimpulzussal megvilágított aranybevonattal ellátott üveg szubsztráton. Az arany, mint nemes [67] átmenetifém, számos alkalmazással rendelkezik a biomedicinától [68], a twistronikán [69] át a hajlékony integrált elektronikáig [70], és az aranybevonattal ellátott tükrök az ultragyors lézeroptika mindennapos elemét képezik. A fém nanofilmekben lezajló ultrarövid elektronemisszió vizsgálatával olyan eredményekre juthatunk, melyek mindezen alkalmazási területek számára fontosak. Az elektron- és a rácshőmérséklet tér- és időbeli fejlődésének meghatározásához megalkottam egy háromdimenziós modellt, amely figyelembe veszi a mintára ferdén fókuszált háromdimenziós lézerimpulzus intenzitásprofilját. A kapcsolódó termionikus emissziós tulajdonságokat a Richardson-Dushman-egyenlet módosított változatának segítségével határozzuk meg a

tér-töltés hatások figyelembe vételével, annak önkonzisztens módon való megoldásával. Meghatároztam továbbá a lézer polarizációjának és beesési szögének szerepét az elektron- és rácshőmérséklet tér-időbeli alakulásában, valamint a termionikus emissziós folyamatokban. Kimutattam a hőmérsékletfüggő reflexió szerepét a lézerenergia elnyelődésében. P-polarizált lézer esetén a maximális felületi elektronhőmérséklet monoton módon emelkedik a lézer beesési szögével, míg S-polarizált esetben ellentétes tendencia figyelhető meg. A termionikus emisszió időtartamát illetően erős függést figyelhetünk meg a a lézer beesési szögétől, valamint ellentétes módon a polarizációtól. Emellett kimutattam a termionikus áram időtartamának erős függését a minta eredendő elektron-rács termalizációs időtartamától is. Az elemzésünkből levont következtetések és meglátások segítenek a fémekben való ultragyors termionemisszió és a kapcsolódó atomi szintű mechanizmusok megértésében és finomhangolásában, valamint a fémekhez hasonlóan viselkedő félvezetőkben is az ultragyors lézerrel való kölcsönhatás során. Ez a tudás termionemissziós áram erősségének és időtartamának növeléséhez vezethet, elősegítve ezzel a hatékony ultragyors termionemissziós emitterek tervezését. Ezekről az eredményekről a [T3] publikált cikkünkben számoltunk be.

4. tézispont

A lézer-anyag kölcsönhatások nemcsak az elektronemissziót befolyásolják. Minden egyes lézerciklus esetében az elektronok ciklus alatti dinamikája is megváltozik a lézer-anyag kölcsönhatás időtartama alatt. Az anyagi válasz pontosabb megértése érdekében meg kell ismernünk ezt a ciklus alatti dinamikát is, ami a magasharmonikus-keltéshez vezethet. Ez a folyamat a lézertér atommal, molekulával vagy periodikus közeggel való nemlineáris kölcsönhatásából ered, és a lézer frekvenciájának egész számú többszörösével rezgő hullámok, felharmonikusok kibocsátását eredményezi. A magasharmonikus-generálás ígéretes technika, mivel képes nagyfrekvenciájú fény keltésére az elektromágneses spektrum extrém ultraibolya és lágy röntgen tartományaiban. Míg az első magasharmonikus-generálást leíró modellek legtöbbször az egy aktív elektron közelítésen vagy az időfüggő Schrödinger-egyenlet megoldásán alapultak, az atomokban történő magasharmonikus-generálás során fellépő többelektronos hatások leírása kiemelkedő jelentőségű, mivel lehetővé teszi a jelenség hátterében álló összetett fizikai folyamatok pontosabb és átfogóbb megértését. Ezért munkámban a többelektronos hatásokat figyelembe vevő időfüggő sűrűségfunkcionál-elméleti megközelítést alkalmazzunk, hogy megvizsgáljuk az ultragyors elektrondinamikát lineárisan polarizált lézerrel, majd kétkörös ellenforgó lézermezőkkel besugárzott Ar atomban. Numerikus megközelítésünket kísérleti eredményekkel validáltam [71], az Ar atom magasharmonikus spektrumának egy fontos jellemzőjének, a Cooper-minimumnak reprodukálásával $\sim 48\ eV$-nál.

A kétszínű ellenforgó cirkulárisan polarizált lézertek olyan környezetet hoznak létre, melyben rendkívül erős és összetett kölcsönhatások játszódnak le az anyaggal. Ez felbecsülhetetlenné teszi őket az alapvető erős térbeli jelenségek, például a magasharmonikus-generáció, a nemlineáris optika, és hasonlóak vizsgálatához. E lézermezők ellenforgó jellege lehetővé teszi az elektronok pályájának és dinamikájának fokozott irányítását. A két mező relatív fázisának és intenzitásának beállításával nagy pontossággal irányíthatjuk és manipulálhatjuk az elektron-kölcsönhatásokat. A legkorszerűbb kísérletek, a félklasszikus analízis és az időfüggő sűrűségfunkcionál-elméleti szimulációk kombinációjával bemutattunk egy hatékony módját a nagyenergiájú és elliptikusan polarizált magasharmonikus sugárzás előállításának és jellemzésének argonból [T4]. Időfüggő sűrűségfunkcionál-elmélet segítségével megmutattam, hogy a másodharmonikus központi hullámhosszának megfelelő hangolásával a magas harmonikus sugárzás központi frekvenciája folytonosan hangolható. Numerikus szimulációink alátámasztják a kísérleti eredményeket, amelyek szerint az argonból származó erősen elliptikus HHG sugárzás $\Delta E \approx 150\ meV$ energiatartományban hangolható $\approx 20\ eV$ spektrális tartományban. Arra számítunk, hogy az intenzív és erősen elliptikus magasharmonikus spektrum hangolásának képessége kiküszöbölheti azokat a korlátokat, amelyeket az izolált cirkulárisan polarizált attoszekundumos impulzusok előállítása során a néhány ciklusos meghajtó impulzusok szükségessége jelent. Ezekről az eredményekről egy, a *Physical Review A* folyóiratban nemrég publikálásra elfogadott cikkünkben számolunk be [T4].

5. tézispont

A kétdimenziós anyagokkal kapcsolatos alapvető ismeretek, és a kapcsolódó előállítási eljárások terén elért előrelépések megnyitották az utat a kétdimenziós anyagok hatékony felhasználása előtt az alkalmazások széles skáláján, a nemlineáris optikától kezdve az optoelektronikán át a fotodetektálásig. Mint már említettük, a foszforén a kétdimenziós anyagok családjának egy viszonylag új, ígéretes tagja, amely jelentős figyelmet kapott a kutatóközösségben olyan különleges tulajdonságai miatt, mint az állítható sávhézag, a magas töltéshordozó mozgékonyság, és a figyelemre méltó síkbeli anizotrópia. Most ezeket az eredendő szerkezeti és elektronikai tulajdonságokat felhasználva vizsgáljuk az ultragyors elektron dinamikát és a magasharmonikus-keltést a lézerrel megvilágított foszforénfelületén. Megvizsgáltuk a külső mechanikai feszültség hatását az elektron-sávszerkezetre és a kapcsolódó nemlineáris hatásokra a foszforénben. Elemzésünk azt mutatja, hogy az ilyen anyagokon belüli erős tér által indukált folyamatok optimalizálhatók és szabályozhatók biaxiális húzó- (+ve) és nyomófeszültség (-ve) alkalmazásával, mely által az elektronszerkezet megfelelően módosítható. A -10%-tól 2%-ig terjedő mechanikai feszültség alkalmazása érdekes eredményeket hoz. A -10%-os feszültség a sávhézag bezáródását idézi elő, míg a 2%-os feszültség a sávhézagot 22%-kal növeli az érintetlen foszforén $0,9\ eV$-os értékéhez képest. Ezek a változások jelentősen befolyásolják a magasharmonikus-keltés hatékonyságát.

A sávhézag feszültség hatására bekövetkező záródása, különösen a 2% és -10% közötti tartományban, hozzájárul a Γ-pont közelében lévő elektronállapot-sűrűség növekedéséhez, ami fokozott elektronikus gerjesztést eredményez. A foszforén saját síkbeli anizotrópiája jelentős tényezőnek bizonyult, ugyanis a harmonikus hozam nagyobb intenzitást mutat, amikor a lézer polarizációja a karosszék-geometria irányában áll, mint amikor a cikk-cakk geometria irányában, minden vizsgált mechanikai feszültség alkalmazása mellett. Az elekton-sávszerkezet külső mechanikai feszültség alkalmazásával való módosítása a gerjesztett elektronok számának növekedését eredményezte, ami végül fokozott magasharmonikus-keltési hatékonysághoz vezet. Különösen szembetűnő a keltési hatásfok közel háromszoros növekedése a karosszék-geometria irányában kifejtett -10%-os mechanikai feszültség esetében. Ezen eredmények feltöltésre kerültek az arXiv repozitórumba [T5], és hamarosan publikációra kerülnek referált folyóiratban. Ez az átfogó tanulmány szélesíti a foszforén kutatásának horizontját, felfedezve eddig feltérképezetlen területeket, és rávilágít annak potenciális alkalmazásaira az extrém-ultraibolya és attoszekundumos nanofotonikában, valamint hatékony asztali nagyharmonikus-keltő forrásként.

A fentiekben tárgyalt legfontosabb eredmények és tézisek fényében a doktori értekezésem fő jelentősége az elektronszerkezeti tulajdonságokra, az elektronemisszióra és az azt követő ultragyors elektrondinamikára gyakorolt hatások vizsgálata és megértése különböző méretű anyagokban, mind lézerimpulzusok terének hatása alatt, mind anélkül. A legkorszerűbb szimulációs megközelítéseken és elméleti modellezésen alapuló eredményeinket összehasonlítjuk és értelmezzük a legfrissebb kísérleti eredményekkel, különös tekintettel azok felhasználhatóságára az alkalmazott tudományban és az anyagmérnöki területeken. Az értekezésben bemutatott átfogó, szimuláción alapuló kutatás javaslatokat tesz a fém- és félvezető nanoszerkezetek foto- és lézerindukált termionikus emissziós tulajdonságainak javítására, amelyeknek olyan széleskörű alkalmazási lehetőségei vannak, mint - csupán néhányat említve - a termionikus emisszión alapuló magas hőmérsékletű napelemek, termionikus energiaátalakítók, vagy nagy fényerejű ultragyors elektron képalkotó eszközök. A disszertációban bemutatott megfelelő optimalizált számítási eszközök és numerikus módszerek segítségével megvizsgáltuk a fémek és kétdimenziós félvezető nanoanyagok fotoindukált termionikus emisszióját, és feltártuk a foszforén és hasonló anyagok alkalmazási lehetőségeit stabil asztali magasharmonikus forrásként. A disszertációmban bemutatott eredményeket számos esetben alátámasztottuk kísérleti eredményekkel, például az analitikus modellünkből származó emissziós áram becslését foszforén esetében. Ezen túlmenően az argonból lineárisan polarizált, valamint kétszínű ellenforgó cirkulárisan polarizált lézertek hatására történő magasharmonikus-keltést is összehasonlítotuk kísérleti eredményekkel, kiegészítve és megmagyarázva a kísérletileg megfigyelt jelenségeket. A disszertációban bemutatott numerikus szimulációkkal célunk a valós idejű lézer-anyag kísérleteket utánozni, így megkerülve a kísérleti kihívásokat. A szimulációk így

a kísérleteket kiegészítő eszközt alkothatnak a fémek, atomok és kétdimenziós anyagok ultragyors elektrondinamikájának megértéséhez a jövőbeli alkalmazásokhoz. Hangsúlyozzuk, hogy a disszertációmban alkalmazott elméleti megközelítések nem korlátozódnak az itt bemutatott anyagokra, hanem más hasonló fémekre/félvezetőkre is kiterjeszthetők. Ebből kifolyólag meggyőződésünk, hogy eredményeink új kísérleti törekvéseket ösztönöznek az ultrarövid elektrondinamika vizsgálatára és a kétdimenziós anyagok későbbi elektronemissziós tulajdonságainak javítására.

Bibliography

[1] Gang Hee Han, Dinh Loc Duong, Dong Hoon Keum, Seok Joon Yun, and Young Hee Lee. "van der Waals Metallic Transition Metal Dichalcogenides". *Chemical Reviews*, 118(13), 2018, pp. 6297–6336. DOI: 10.1021/acs.chemrev.7b00618.

[2] Subhamoy Ghatak, Atindra Nath Pal, and Arindam Ghosh. "Nature of Electronic States in Atomically Thin MoS_2 Field-Effect Transistors". *ACS Nano*, 5(10), 2011, pp. 7707–7712. DOI: 10.1021/nn202852j.

[3] Steve M. Young and Charles L. Kane. "Dirac Semimetals in Two Dimensions". *Physical Review Letters*, 115(12), 2015. DOI: 10.1103/physrevlett.115.126803.

[4] K. Ozdogan, M. Upadhyay Kahaly, H. N. Alshareef, and U. Schwingenschlögl. "Anomalous enhancement of the thermoelectric figure of merit by V co-doping of Nb-SrTiO3". *Applied Physics Letters*, 100(19), 2012. DOI: 10.1063/1.4714541.

[5] Eleftherios Goulielmakis, Zhi-Heng Loh, Adrian Wirth, Robin Santra, Nina Rohringer, Vladislav S. Yakovlev, Sergey Zherebtsov, Thomas Pfeifer, Abdallah M. Azzeer, Matthias F. Kling, Stephen R. Leone, and Ferenc Krausz. "Real-time observation of valence electron motion". *Nature*, 466(7307), 2010, pp. 739–743. DOI: 10.1038/nature09212.

[6] M. Upadhyay Kahaly, S. Nazir, and U. Schwingenschlögl. "Band structure engineering and vacancy induced metallicity at the GaAs-AlAs interface". *Applied Physics Letters*, 99(12), 2011. DOI: 10.1063/1.3643049.

[7] M. Upadhyay Kahaly, Shikha Misra, and S. K. Mishra. "Photo-assisted electron emission from illuminated monolayer graphene". *Journal of Applied Physics*, 121(20), 2017, p. 205110. DOI: 10.1063/1.4984318.

[8] Shijing Tan, Adam Argondizzo, Cong Wang, Xuefeng Cui, and Hrvoje Petek. "Ultrafast Multiphoton Thermionic Photoemission from Graphite". *Physical Review X*, 7(1), 2017. DOI: 10.1103/physrevx.7.011004.

[9] Sha Li and R. R. Jones. "High-energy electron emission from metallic nano-tips driven by intense single-cycle terahertz pulses". *Nature Communications*, 7(1), 2016. DOI: 10.1038/ncomms13405.

[10] Young Duck Kim, Yuanda Gao, Ren-Jye Shiue, Lei Wang, Ozgur Burak Aslan, Myung-Ho Bae, Hyungsik Kim, Dongjea Seo, Heon-Jin Choi, Suk Hyun Kim, Andrei Nemilentsau, Tony Low, Cheng Tan, Dmitri K. Efetov, Takashi Taniguchi, Kenji Watanabe, Kenneth L. Shepard, Tony F. Heinz, Dirk Englund, and James Hone. "Ultrafast Graphene Light Emitters". *Nano Letters*, 18(2), 2018, pp. 934–940. DOI: 10.1021/acs.nanolett.7b04324.

[11] Gongtao Wu, Xianlong Wei, Song Gao, Qing Chen, and Lianmao Peng. "Tunable graphene micro-emitters with fast temporal response and controllable electron emission". *Nature Communications*, 7(1), 2016. DOI: 10.1038/ncomms11513.

[12] Mousumi Upadhyay Kahaly and Umesh V. Waghmare. "Contrast in the Electronic and Magnetic Properties of Doped Carbon and Boron Nitride Nanotubes: A First-Principles Study". *The Journal of Physical Chemistry C*, 112(10), 2008, pp. 3464–3472. DOI: 10.1021/jp072340d.

[13] Matthew P. McDonald, Rusha Chatterjee, Jixin Si, Boldizsár Jankó, and Masaru Kuno. "Dimensional crossover in semiconductor nanostructures". *Nature Communications*, 7(1), 2016. DOI: 10.1038/ncomms12726.

[14] Melanie Müller, Alexander Paarmann, and Ralph Ernstorfer. "Femtosecond electrons probing currents and atomic structure in nanomaterials". *Nature Communications*, 5(1), 2014. DOI: 10.1038/ncomms6292.

[15] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov. "Electric Field Effect in Atomically Thin Carbon Films". *Science*, 306(5696), 2004, pp. 666–669. DOI: 10.1126/science.1102896.

[16] S. Madas, S. K. Mishra, S. Kahaly, and M. Upadhyay Kahaly. "Superior Photo-thermionic electron Emission from Illuminated Phosphorene Surface". *Scientific Reports*, 9(1), 2019. DOI: 10.1038/s41598-019-44823-x.

[17] D. Pacilé, J. C. Meyer, Ç. Ö. Girit, and A. Zettl. "The two-dimensional phase of boron nitride: Few-atomic-layer sheets and suspended membranes". *Applied Physics Letters*, 92(13), 2008. DOI: 10.1063/1.2903702.

[18] Pranab Kumar Das, D. Di Sante, I. Vobornik, J. Fujii, T. Okuda, E. Bruyer, A. Gyenis, B. E. Feldman, J. Tao, R. Ciancio, G. Rossi, M. N. Ali, S. Picozzi, A. Yadzani, G. Panaccione, and R. J. Cava. "Layer-dependent quantum cooperation of electron and hole states in the anomalous semimetal WTe_2". *Nature Communications*, 7(1), 2016. DOI: 10.1038/ncomms10847.

[19] Yu Saito, Tsutomu Nojima, and Yoshihiro Iwasa. "Highly crystalline 2D superconductors". *Nature Reviews Materials*, 2(1), 2016. DOI: 10.1038/natrevmats.2016.94.

[20] Guorui Chen, Lili Jiang, Shuang Wu, Bosai Lyu, Hongyuan Li, Bheema Lingam Chittari, Kenji Watanabe, Takashi Taniguchi, Zhiwen Shi, Jeil Jung, Yuanbo Zhang, and Feng Wang. "Evidence of a gate-tunable Mott insulator in a trilayer graphene moiré superlattice". *Nature Physics*, 15(3), 2019, pp. 237–241. DOI: 10.1038/s41567-018-0387-2.

[21] Xiaomeng Liu, Kenji Watanabe, Takashi Taniguchi, Bertrand I. Halperin, and Philip Kim. "Quantum Hall drag of exciton condensate in graphene". *Nature Physics*, 13(8), 2017, pp. 746–750. DOI: 10.1038/nphys4116.

[22] Qing Hua Wang, Kourosh Kalantar-Zadeh, Andras Kis, Jonathan N. Coleman, and Michael S. Strano. "Electronics and optoelectronics of two-dimensional transition metal dichalcogenides". *Nature Nanotechnology*, 7(11), 2012, pp. 699–712. DOI: 10.1038/nnano.2012.193.

[23] A. N. Grigorenko, M. Polini, and K. S. Novoselov. "Graphene plasmonics". *Nature Photonics*, 6(11), 2012, pp. 749–758. DOI: 10.1038/nphoton.2012.262.

[24] Saptarshi Das, Hong-Yan Chen, Ashish Verma Penumatcha, and Joerg Appenzeller. "High Performance Multilayer MoS_2 Transistors with Scandium Contacts". *Nano Letters*, 13(1), 2012, pp. 100–105. DOI: 10.1021/nl303583v.

[25] Kelvin J. A. Ooi and Dawn T. H. Tan. "Nonlinear graphene plasmonics". *Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences*, 473(2206), 2017, p. 20170433. DOI: 10.1098/rspa.2017.0433.

[26] Hassan A. Hafez, Sergey Kovalev, Klaas-Jan Tielrooij, Mischa Bonn, Michael Gensch, and Dmitry Turchinovich. "Terahertz Nonlinear Optics of Graphene: From Saturable Absorption to High-Harmonics Generation". *Advanced Optical Materials*, 8(3), 2019, p. 1900771. DOI: 10.1002/adom.201900771.

[27] Fengnian Xia, Han Wang, Di Xiao, Madan Dubey, and Ashwin Ramasubramaniam. "Two-dimensional material nanophotonics". *Nature Photonics*, 8(12), 2014, pp. 899–907. DOI: 10.1038/nphoton.2014.271.

[28] Zhipei Sun, Amos Martinez, and Feng Wang. "Optical modulators with 2D layered materials". *Nature Photonics*, 10(4), 2016, pp. 227–238. DOI: 10.1038/nphoton.2016.15.

[29] Francesco Bonaccorso, Luigi Colombo, Guihua Yu, Meryl Stoller, Valentina Tozzini, Andrea C. Ferrari, Rodney S. Ruoff, and Vittorio Pellegrini. "Graphene, related two-dimensional crystals, and hybrid systems for energy conversion and storage". *Science*, 347(6217), 2015. DOI: 10.1126/science.1246501.

[30] Babak Anasori, Maria R. Lukatskaya, and Yury Gogotsi. "2D metal carbides and nitrides (MXenes) for energy storage". *Nature Reviews Materials*, 2(2), 2017. DOI: 10.1038/natrevmats.2016.98.

[31] Karim Khan, Ayesha Khan Tareen, Muhammad Aslam, Yupeng Zhang, Renheng Wang, Zhengbiao Ouyang, Zhongyi Gou, and Han Zhang. "Recent advances in two-dimensional materials and their nanocomposites in sustainable energy conversion applications". *Nanoscale*, 11(45), 2019, pp. 21622–21678. DOI: 10.1039/c9nr05919a.

[32] John R. Schaibley, Hongyi Yu, Genevieve Clark, Pasqual Rivera, Jason S. Ross, Kyle L. Seyler, Wang Yao, and Xiaodong Xu. "Valleytronics in 2D materials". *Nature Reviews Materials*, 1(11), 2016. DOI: 10.1038/natrevmats.2016.55.

[33] Stephan Roche, Johan Åkerman, Bernd Beschoten, Jean-Christophe Charlier, Mairbek Chshiev, Saroj Prasad Dash, Bruno Dlubak, Jaroslav Fabian, Albert Fert, Marcos Guimarães, Francisco Guinea, Irina Grigorieva, Christian Schönenberger, Pierre Seneor, Christoph Stampfer, Sergio O Valenzuela, Xavier Waintal, and Bart van Wees. "Graphene spintronics: the European Flagship perspective". *2D Materials*, 2(3), 2015, p. 030202. DOI: 10.1088/2053-1583/2/3/030202.

[34] Yuan Cao, Valla Fatemi, Shiang Fang, Kenji Watanabe, Takashi Taniguchi, Efthimios Kaxiras, and Pablo Jarillo-Herrero. "Unconventional superconductivity in magic-angle graphene superlattices". *Nature*, 556(7699), 2018, pp. 43–50. DOI: 10.1038/nature26160.

[35] Vinod K. Sangwan and Mark C. Hersam. "Neuromorphic nanoelectronic materials". *Nature Nanotechnology*, 15(7), 2020, pp. 517–528. DOI: 10.1038/s41565-020-0647-z.

[36] Toan Trong Tran, Kerem Bray, Michael J. Ford, Milos Toth, and Igor Aharonovich. "Quantum emission from hexagonal boron nitride monolayers". *Nature Nanotechnology*, 11(1), 2015, pp. 37–41. DOI: 10.1038/nnano.2015.242.

[37] Kenneth Lopata and Niranjan Govind. "Modeling Fast Electron Dynamics with Real-Time Time-Dependent Density Functional Theory: Application to Small Molecules and Chromophores". *Journal of Chemical Theory and Computation*, 7(5), 2011, pp. 1344–1355. DOI: 10.1021/ct200137z.

[38] Mousumi Upadhyay Kahaly, Saibabu Madas, Boris Mesits, and Subhendu Kahaly. "Tunable ultrafast thermionic emission from femtosecond-laser hot spot on a metal surface by control of laser polarization and angle of incidence: A numerical investigation". *Applied Surface Science*, 643, 2024, p. 158668. DOI: 10.1016/j.apsusc.2023.158668.

[39] Shambhu Ghimire and David A. Reis. "High-harmonic generation from solids". *Nature Physics*, 15(1), 2018, pp. 10–16. DOI: 10.1038/s41567-018-0315-5.

[40] T. P. Kaloni, M. Upadhyay Kahaly, Y. C. Cheng, and U. Schwingenschlögl. "Mechanism of Si intercalation in defective graphene on SiC". *Journal of Materials Chemistry*, 22(44), 2012, p. 23340. DOI: 10.1039/c2jm35127g.

[41] Mousumi Upadhyay Kahaly. "Defect states in carbon nanotubes and related band structure engineering: A first-principles study". *Journal of Applied Physics*, 105(2), 2009. DOI: 10.1063/1.3072695.

[42] Y. S. Ang and L. K. Ang. "Current-Temperature Scaling for a Schottky Interface with Nonparabolic Energy Dispersion". *Physical Review Applied*, 6(3), 2016. DOI: 10.1103/physrevapplied.6.034013.

[43] Shikha Misra, S. K. Mishra, and M. S. Sodha. "Photoelectric sheath formation around small spherical objects in space". *Physics of Plasmas*, 22(4), 2015, p. 043705. DOI: 10.1063/1.4918946.

[44] Shikha Misra, M. Upadhyay Kahaly, and S. K. Mishra. "Thermionic emission from monolayer graphene, sheath formation and its feasibility towards thermionic converters". *Journal of Applied Physics*, 121(6), 2017, p. 065102. DOI: 10.1063/1.4975788.

[45] Han Liu, Adam T. Neal, Zhen Zhu, Zhe Luo, Xianfan Xu, David Tománek, and Peide D. Ye. "Phosphorene: An Unexplored 2D Semiconductor with a High Hole Mobility". *ACS Nano*, 8(4), 2014, pp. 4033–4041. DOI: 10.1021/nn501226z.

[46] P. Hohenberg and W. Kohn. "Inhomogeneous Electron Gas". *Physical Review*, 136(3B), 1964, B864–B871. DOI: 10.1103/physrev.136.b864.

[47] Paolo Giannozzi et al. "QUANTUM ESPRESSO: a modular and open-source software project for quantum simulations of materials". *Journal of Physics: Condensed Matter*, 21(39), 2009, p. 395502. DOI: 10.1088/0953-8984/21/39/395502.

[48] A. Block, M. Liebel, R. Yu, M. Spector, Y. Sivan, F. J. García de Abajo, and N. F. van Hulst. "Tracking ultrafast hot-electron diffusion in space and time by ultrafast thermomodulation microscopy". *Science Advances*, 5(5), 2019, eaav8965. DOI: 10.1126/sciadv.aav8965.

[49] Zhongyang Wang, Zhengquan Zhang, Zhizhan Xu, and Qiang Lin. "Space-time profiles of an ultrashort pulsed Gaussian beam". *IEEE Journal of Quantum Electronics*, 33(4), 1997, pp. 566–573. DOI: 10.1109/3.563385.

[50] S.Y. Wang, Y. Ren, C.W. Cheng, J.K. Chen, and D.Y. Tzou. "Micromachining of copper by femtosecond laser pulses". *Applied Surface Science*, 265, 2013, pp. 302–308. DOI: 10.1016/j.apsusc.2012.10.200.

[51] S.-S. Wellershoff, J. Hohlfeld, J. Güdde, and E. Matthias. "The role of electron-phonon coupling in femtosecond laser damage of metals". *Applied Physics A Materials Science & Processing*, 69(S1), 1999, S99–S107. DOI: 10.1007/s003399900305.

[52] Max Born and Emil Wolf. "Basic properties of the electromagnetic field". *Principles of optics*, 44, 1980, pp. 1–70.

[53] A. Vial and T. Laroche. "Comparison of gold and silver dispersion laws suitable for FDTD simulations". *Applied Physics B*, 93(1), 2008, pp. 139–143. DOI: 10.1007/s00340-008-3202-4.

[54] M Born and E Wolf. *Principles of Optics*. Pergamon Press Ltd., Oxford., 1964.

[55] P. G. Etchegoin, E. C. Le Ru, and M. Meyer. "An analytic model for the optical properties of gold". *The Journal of Chemical Physics*, 125(16), 2006, p. 164705. DOI: 10.1063/1.2360270.

[56] Neil W Ashcroft and N David Mermin. "Introduction to Solid State Physics". *NY: Brooks Cole*, 1976.

[57] J. Leng, J. Opsal, H. Chu, M. Senko, and D.E. Aspnes. "Analytic representations of the dielectric functions of materials for device and structural modeling". *Thin Solid Films*, 313-314, 1998, pp. 132–136. DOI: 10.1016/s0040-6090(97)00799-2.

[58] Andrew N. Smith and Pamela M. Norris. "Influence of intraband transitions on the electron thermoreflectance response of metals". *Applied Physics Letters*, 78(9), 2001, pp. 1240–1242. DOI: 10.1063/1.1351523.

[59] P. G. Etchegoin, E. C. Le Ru, and M. Meyer. "Erratum: "An analytic model for the optical properties of gold" [J. Chem. Phys. 125, 164705 (2006)]". *The Journal of Chemical Physics*, 127(18), 2007, p. 189901. DOI: 10.1063/1.2802403.

[60] P. B. Johnson and R. W. Christy. "Optical Constants of the Noble Metals". *Physical Review B*, 6(12), 1972, pp. 4370–4379. DOI: 10.1103/physrevb.6.4370.

[61] K. Yabana and G. F. Bertsch. "Time-dependent local-density approximation in real time". *Physical Review B*, 54(7), 1996, pp. 4484–4487. DOI: 10.1103/physrevb.54.4484.

[62] Xi Chu and Shih-I Chu. "Time-dependent density-functional theory for molecular processes in strong fields: Study of multiphoton processes and dynamical response of individual valence electrons of N_2 in intense laser fields". *Physical Review A*, 64(6), 2001. DOI: 10.1103/physreva.64.063404.

[63] K. Burnett, V. C. Reed, J. Cooper, and P. L. Knight. "Calculation of the background emitted during high-harmonic generation". *Physical Review A*, 45(5), 1992, pp. 3347–3349. DOI: 10.1103/physreva.45.3347.

[64] Xi Liu, Xiaosong Zhu, Liang Li, Yang Li, Qingbin Zhang, Pengfei Lan, and Peixiang Lu. "Selection rules of high-order-harmonic generation: Symmetries of molecules and laser fields". *Physical Review A*, 94(3), 2016. DOI: 10.1103/physreva.94.033410.

[65] Nicolas Tancogne-Dejean, Oliver D. Mücke, Franz X. Kärtner, and Angel Rubio. "Impact of the Electronic Band Structure in High-Harmonic Generation Spectra of Solids". *Physical Review Letters*, 118(8), 2017. DOI: 10.1103/physrevlett.118.087403.

[66] M. Upadhyay Kahaly and U. Schwingenschlögl. "Enhanced solar light absorption of graphene by interaction with anisole". *Carbon*, 77, 2014, pp. 76–79. DOI: 10.1016/j.carbon.2014.05.005.

[67] Maria Balcerzak. *Noble Metals, Analytical Chemistry of*. 2021. DOI: 10.1002/9780470027318.a2411.pub3.

[68] Min Xu, Yiling Song, Jinping Wang, and Nan Li. "Anisotropic transition metal–based nanomaterials for biomedical applications". *VIEW*, 2(4), 2021, p. 20200154. DOI: 10.1002/viw.20200154.

[69] Max Heyl and Emil J. W. List-Kratochvil. "Only gold can pull this off: mechanical exfoliations of transition metal dichalcogenides beyond scotch tape". *Applied Physics A*, 129(1), 2022. DOI: 10.1007/s00339-022-06297-z.

[70] Masahito Takakuwa, Kenjiro Fukuda, Tomoyuki Yokota, Daishi Inoue, Daisuke Hashizume, Shinjiro Umezu, and Takao Someya. "Direct gold bonding for flexible integrated electronics". *Science Advances*, 7(52), 2021. DOI: 10.1126/sciadv.abl6228.

[71] C.-G. Wahlström, J. Larsson, A. Persson, T. Starczewski, S. Svanberg, P. Salières, Ph. Balcou, and Anne L'Huillier. "High-order harmonic generation in rare gases with an intense short-pulse laser". *Physical Review A*, 48(6), 1993, pp. 4709–4720. DOI: 10.1103/physreva.48.4709.

Own publications

[T1] **Saibabu Madas**, S. K. Mishra, and Mousumi Upadhyay Kahaly. "Enhanced electron emission from coated metal targets: Effect of surface thickness on performance". *AIP Advances*, 8(3), 2018, p. 035019. DOI: 10.1063/1.5012861. **IF: 1.6**

[T2] **Saibabu Madas**, S. K. Mishra, S. Kahaly, and M. Upadhyay Kahaly. "Superior Photo-thermionic electron Emission from Illuminated Phosphorene Surface". *Scientific Reports*, 9(1), 2019. DOI: 10.1038/s41598-019-44823-x. **IF: 4.9**

[T3] Mousumi Upadhyay Kahaly, **Saibabu Madas**, Boris Mesits, and Subhendu Kahaly. "Tunable ultrafast thermionic emission from femtosecond-laser hot spot on a metal surface by control of laser polarization and angle of incidence: A numerical investigation". *Applied Surface Science*, 643, 2024, p. 158668. DOI: 10.1016/j.apsusc.2023.158668. **IF: 6.7**

[T4] E. Vassakis, **Saibabu Madas**, L. Spachis, T. Lamprou, I. Orfanos, S. Kahaly, M. Upadhyay Kahaly, D. Charalambidis, and E. Skantzakis. "Energetic, tunable, highly elliptically polarized higher harmonics generated by intense two-color counter-rotating laser fields". *Physical Review A*, 2023. **IF: 2.9. Just accepted**

[T5] **Saibabu Madas**, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. *Controlling high-harmonic generation from strain engineered monolayer phosphorene*. 2023.

Other scientific publications

[C1] Manju, Megha Jain, **Saibabu Madas**, Pargam Vashishtha, Parasmani Rajput, Govind Gupta, Mousumi Upadhyay Kahaly, Kemal Özdoğan, Ankush Vij, and Anup Thakur. "Oxygen vacancies induced photoluminescence in $SrZnO_2$ nanophosphors probed by theoretical and experimental analysis". *Scientific Reports*, 10(1), 2020. DOI: 10.1038/s41598-020-74436-8. **IF: 4.9**

IF: Impact factor
Cumulative impact factor: 21
MTMT number: 10082058

Conference presentations

*Presenting author; OP- Oral Presentation; PP- Poster presentation

[OP1] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Superior Photo-thermionic Electron Emission from Illuminated Phosphorene Surface". *In EUCALL Joint Foresight Topical Workshop: Theory and Simulation of Photon-Matter Interaction, Szeged, Hungary*, 2018.

[OP2] **Saibabu Madas***, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Harmonizing light and strain: piezotronic control of high harmonic generation from tunable monolayer phosphorene". *In Annual meeting of the International Max Planck Research School on Advanced Photon Science (IMPRS-APS), Vienna, Austria*, 2023.

[PP1] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Enhanced Photo-Electron Emission from Illuminated Phosphorene Surface". *In Joint Training School of the COST actions CM1401 Our Astrochemical History & CM1405 MOLIM: Molecules in Motion New avenues in molecular theories: From the lab to beyond the Earth, Belgrade, Serbia*, 2017.

[PP2] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Enhanced Photo-Electron Emission from Illuminated Phosphorene Surface". *In 14th International Conference on Multiphonon processes, Budapest, Hungary*, 2017.

[PP3] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Photo-Electron and Thermionic Emission from an Illuminated Phosphorene Surface". *In Psi-k workshop on 2D layered materials for opto-electronics: a theoretical/computational perspective, Rome, Italy*, 2017.

[PP4] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Photo-Electron and Thermionic Emission from an Illuminated Phosphorene Surface". *In Winter College on Optics: Extreme Nonlinear Optics, Attosecond Science and High-field Physics, Trieste, Italy*, 2018.

[PP5] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Photo-induced ultrafast electron emission and photonic bandgap engineering in phosphorene". *In 7th International Conference on Attosecond Science and Technology, Szeged, Hungary*, 2019.

[PP6] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Photo-induced ultrafast electron emission and photonic bandgap engineering in phosphorene". *In 1^{st} Training School of the COST Action CA17126 - TUMIEE (Towards Understanding and Modelling Intense Electronic Excitation), Crete, Greece*, 2019.

[PP7] **Saibabu Madas***, S. K. Mishra, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Photo-induced ultrafast electron emission and photonic bandgap engineering in phosphorene". *In ICFO school on "The Frontiers of Light: Attosecond science and extreme photonics, Barcelona, Spain*, 2019.

[PP8] A. Antunes*, M. Hussain, **Saibabu Madas**, Mousumi Upadhyay Kahaly, Subhendu Kahaly, M. Fajardo, and G. Williams. "Shining Light on Solids: A TDDFT study of Solid-State HHG". *In ELI Summer School, Szeged, Hungary*, 2022.

[PP9] A. Antunes*, M. Hussain, **Saibabu Madas**, Mousumi Upadhyay Kahaly, Subhendu Kahaly, M. Fajardo, and G. Williams. "Shining Light on Solids: A TDDFT study of Solid-State HHG". *In SPIE Optics + Optoelectronics, Prague, Czech Republic*, 2023.

[PP10] **Saibabu Madas***, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Controlling High-Harmonic Generation from Strain-Engineered Monolayer Quantum Flatland". *In ELI Summer School, Dolní Břežany, Czech Republic*, 2023.

[PP11] **Saibabu Madas***, Arjun Nayak Puttur, Subhendu Kahaly, and Mousumi Upadhyay Kahaly. "Tuning high-harmonic generation from ultrathin monolayer black phosphorous by controlled strain engineering". *In 9th International Conference on Attosecond Science and Technology, Jeju island, Korea*, 2023.

Declaration of Co-authorship

I, Dr. Subhendu Kahaly hereby declare that as a co-author I recognize the thesis points 2 to 4 for Ref. [1-3] given below as individual result by Saibabu Madas and declare that I will and did not use these results to obtain any scientific degree.

[1] S. Madas, S. K. Mishra, S. Kahaly, and M. Upadhyay Kahaly. "Superior Photo- thermionic electron Emission from Illuminated Phosphorene Surface". Scientific Reports, 9(1), 2019. doi: 10.1038/s41598-019-44823-x.

[2] M. Upadhyay Kahaly, S. Madas, B. Mesits, and S. Kahaly. "Tunable ultrafast thermionic emission from femtosecond-laser hot spot on a metal surface by control of laser polarization and angle of incidence: A numerical investigation". Applied Surface Science, 643, 2024, p. 158668. doi: 10.1016/j.apsusc.2023.158668.

[3] E. Vassakis, S. Madas, L. Spachis, T. Lamprou, I. Orfanos, S. Kahaly, M. Upadhyay Kahaly, D. Charalambidis, and E. Skantzakis. "Energetic, tunable, highly elliptically polarized higher harmonics generated by intense two-color counter-rotating laser fields". Physical Review A, 2023. Just accepted

Date and place

Signature

Declaration of Co-authorship

I, Dr. Mousumi Upadhyay Kahaly hereby declare that as a co-author I recognize the thesis points 1 to 4 for Ref. [1-4] given below as individual result by Saibabu Madas and declare that I will and did not use these results to obtain any scientific degree.

[1] S. Madas, S. K. Mishra, and Mousumi Upadhyay Kahaly. "Enhanced electron emission from coated metal targets: Effect of surface thickness on performance". AIP Advances, 8(3), 2018, p. 035019. doi: 10.1063/1.5012861.

[2] S. Madas, S. K. Mishra, S. Kahaly, and M. Upadhyay Kahaly. "Superior Photo- thermionic electron Emission from Illuminated Phosphorene Surface". Scientific Reports, 9(1), 2019. doi: 10.1038/s41598-019-44823-x.

[3] M. Upadhyay Kahaly, S. Madas, B. Mesits, and S. Kahaly. "Tunable ultrafast thermionic emission from femtosecond-laser hot spot on a metal surface by control of laser polarization and angle of incidence: A numerical investigation". Applied Surface Science, 643, 2024, p. 158668. doi: 10.1016/j.apsusc.2023.158668.

[4] E. Vassakis, S. Madas, L. Spachis, T. Lamprou, I. Orfanos, S. Kahaly, M. Upadhyay Kahaly, D. Charalambidis, and E. Skantzakis. "Energetic, tunable, highly elliptically polarized higher harmonics generated by intense two-color counter-rotating laser fields". Physical Review A, 2023. Just accepted

Date and place

Signature