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研究課題名（和文）Ultrafast femtosecond laser control of electron dynamics in two-dimensional strong spin-orbit coupling materials

研究課題名（英文）Ultrafast femtosecond laser control of electron dynamics in two-dimensional strong spin-orbit coupling materials

研究代表者
Ｈａｓｈｍｉ　Ａｒｑｕｍ（Hashmi, Arqum)

東京大学・大学院工学系研究科（工学部）・特任研究員

研究者番号：９０８１５３２５

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研究成果の概要（和文）：二次元単分子膜は、強い光物質相互作用と超高速広帯域光学応答を特徴とする新しい分野となっている。この目的のために、我々は、超高速時間依存電子ダイナミクスを研究するためのベンチマークとして、TDDFTとマクスウェル方程式の形式を用いた。SOCを組み込むことによって、我々の研究は、二次元物質における電子ダイナミクスのさまざまな側面を探求している。我々は、電子ダイナミクスをサブフェムト秒スケールで制御できる二次元半導体と半金属を研究している。これにより、超短パルスでキャリアダイナミクスを制御し、超高速情報処理のためのフェムト秒スケールのスピントロニクスを探求する機会が開かれる。

研究成果の学術的意義や社会的意義
The scientific importance of this research is about understanding how electrons move really fast. It helps us learn how materials behave when interact with light. This can be useful for making new technologies like faster computers and communication devices and improved ways to store information.

研究成果の概要（英文）：Owing to their extraordinary physical properties, 2D monolayers have become an emerging field, featuring strong light-matter interactions and ultrafast broadband optical responses. A comprehensive computational framework to explore and explain the different aspects of ultrafast dynamics is urgently needed.
To address this, we used the formalism of TDDFT with Maxwell equations as a benchmark to study ultrafast time-dependent electron dynamics. By incorporating spin-orbit couplings, our study explores various aspects of electron dynamics in 2D materials. We examined 2D semiconductors and semimetals, which can control electron dynamics on the sub-femtosecond timescale, faster than electron-electron (tens of fs) and electron-phonon scattering (hundreds of fs). We found the possibility to control carrier dynamics up to the femtosecond timescale through ultrashort pulses and explored spintronics at the femtosecond scale, opening opportunities for extremely fast information processing.

研究分野：Condensed matter physics

キーワード：Ultrafast　TDDFT　Nonlinear optics　Spin-orbit coupling　femtosecond

様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９（共通）

１．研究開始当初の背景

Within a wide class of 2D materials family, monolayers with strong spin-orbit coupling (SOC) are getting special attention because SOC lifts the spin degeneracy without any external field. In this regard, two different categories of 2D materials

(i) Transition metal dichalcogenides (TMDC-materials) with broken inversion symmetry and

(ii) Topological insulators such as Bi_2Se_3 where the bulk is semiconducting and the surfaces are semi-metallic presents an exotic state of matter, such as spin-valley locking, helical spin structure, chiral anomaly, and dissipation-less currents.

The research on TMDC and topological insulators thin layers has been revolved around the electron-phonon interactions, excited electron relaxation times and other optical responses that are typically in the range of pico (10^{-12}) to nano (10^{-9}) second. On the other hand, very little attention is given to strong electromagnetic fields which can open the possibility of controlling electron dynamics on the fs or sub-fs timescale. Nonetheless, the fs laser control of electron dynamics in 2D strong SOC materials largely remains elusive. Strong-field electron dynamics is a complex phenomenon due to the involvement of intraband and interband transitions, electron-hole couplings, and electron correlation which also depend on crystal symmetries and laser polarization. Thus, a comprehensive theoretical and computational framework to explore and explain the different aspects of ultrafast dynamics is in urgent need which is missing right now.

２．研究の目的

Through this study, I intend to focus on these key issues,

(i) How the shape of the incident electric field pulse (linear, circular, and asymmetric electric field) controlled by the carrier-envelope phase (CEP) affect the charge and spin dynamics in the presence of SOC.

(ii) How field strength affects the electron dynamics in the linear and nonlinear optics limits

The characteristics and applications of 2D materials with ultrashort pulses are quite diverse in the presence of strong SOC. In this regard, comprehensive theoretical and computational studies for the explanation of underlying physics of different aspects of spintronics in WSe_2 and Bi_2Se_3 due to SOC are lacking. These physical aspects of electron spin are important as they interplay with the symmetries and incident light polarization. Furthermore, understanding the roles of these physical aspects in 2D TMDs and topological insulators can accelerate the experimental research in the field of ultrafast opto-spintronics.

３．研究の方法

We employ real-time-dependent density functional theory (TDDFT) +Maxwell's equation approach, which is known for its efficiency in effectively describing linear and nonlinear electron dynamics. To fully incorporate the spin-orbit coupling effect and correct description of charge carriers, the noncollinear version of TDDFT is used. The theoretical formalism of the time-dependent Kohn-Sham (TDKS) method and maxwell equations with SOC has already been implemented in SALMON.

Electron dynamics in a unit cell:

TDKS equation for the Bloch orbital is written as,

$$i\hbar \frac{\partial}{\partial t} u_{b,k}(r,t) = \left[\frac{1}{2m}\left(-i\hbar\nabla + \frac{1}{c} A(t) + \hbar k\right)^2 - e\varphi(r,t) + v_{ion} + v_{XC}\right] u_{b,k}(r,t) \qquad (1)$$

By using $u_{b,k}(r,t)$, one can calculate the electric current density $J(t)$, which is averaged over the unit cell as

$$J(t) = \frac{-e}{m}\int_{\Omega} \frac{dr}{\Omega} \sum_{bk}^{occ} u_{b,k}(r,t)^* \times \left(-i\hbar\nabla + \frac{1}{c} A(t) + \hbar k\right) u_{b,k}(r,t) + \delta J(t) \qquad (2)$$

Propagation of electromagnetic fields:

Maxwell equations describe the propagation of electromagnetic fields in the form of the vector potential $A_z(t)$ described as,

$$\frac{1}{c^2}\frac{\partial^2 A_z(t)}{\partial t^2} - \frac{\partial^2 A_z(t)}{\partial Z^2} = \frac{4\pi}{c} J_z(t) \quad (3)$$

Thus, the TDKS Eq. (1) becomes as follows,

$$i\hbar \frac{\partial}{\partial t} u_{b,k}(r,t) = \left[\frac{1}{2m}(-i\hbar\nabla + \hbar k + \frac{e}{c} A_z(t))^2 - e\varphi(r,t) + v_{ion} + v_{XC}\right] u_{b,k}(r) \quad (4)$$

４．研究成果

Regarding the ultrafast electron dynamics in 2D layers of TMDC. I investigated the carrier dynamics in terms of valley polarization in WSe_2 monolayer via single and two-color laser field. The valley selection rules suggest that linearly polarized light couples equally to both valleys and valley polarization cannot be achieved by a linearly polarized field. I aimed to induce valley asymmetry by linearly polarized pulses which are considered not achievable by a linearly polarized field. It is possible to create an asymmetric laser field by mixing a fundamental frequency and its second harmonic. The two pulses intensity ratio and the relative carrier-envelope phase (CEP) control the valley polarization. I compared the valley polarization results of two-color laser scheme results with the single-color pulse. I found that the valley polarization via two-color control exceeds the single-color scheme by 1.2 times as the two-colors field exhibits more asymmetry in its temporal shape. The results reveal a convenient new path toward the optical control of valley pseudospins.

On the study of topological insulator, the linear and nonlinear optical response of a topological insulator Bi_2Se_3 is investigated. The electron dynamics along with the saturable absorption and high harmonic generation are examined at weak and strong laser fields. This theoretical study provides insight into the weak and strong-field-driven dynamics of Dirac-Fermions in the ultrathin films of the topological insulators. I have explored the bulk and surface state by weak and strong field response in Bulk and in 3-layered system of Bi_2Se_3. Our results demonstrate that a linearly polarized resonant pulse generates higher order odd harmonics by strong field. The strength of harmonics in bulk Bi_2Se_3 is higher than in the layered system.

５．主な発表論文等

〔雑誌論文〕　計0件

〔学会発表〕　計5件（うち招待講演　0件／うち国際学会　4件）

１．発表者名 Hashmi Arqum
２．発表標題 Weak and Strong Field Control of Valley Polarization in WSe2 Monolayer
３．学会等名 Conference on Lasers and Electro-Optics（国際学会）
４．発表年 2022年

１．発表者名 Hashmi Arqum
２．発表標題 Optical Field Control of Electron Dynamics in WSe2 Monolayer
３．学会等名 15th Pacific Rim Conference on Lasers and Electro-Optics（国際学会）
４．発表年 2022年

１．発表者名 Hashmi Arqum
２．発表標題 Sub-cycle control of valley polarization in WSe2 monolayer
３．学会等名 8th International Conference on Attosecond Science and Technology（国際学会）
４．発表年 2022年

１．発表者名 Hashmi Arqum
２．発表標題 Valley selective excitations in WSe2 monolayer by (ω+2ω) pulses
３．学会等名 83rd JSAP Autumn Meeting
４．発表年 2022年

１．発表者名 Hashmi Arqum
２．発表標題 Valley polarization in WSe2 monolayer: CEP control of ω and ω+2ω Pulses
３．学会等名 9th Workshop on Time-Dependent Density-Functional Theory: Prospects and Applications（国際学会）
４．発表年 2022年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

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６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関