

Functional Materials Science and Engineering Research Section

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1. Introduction

Our research section focuses on the physical properties, functions, and energy applications of quantum materials that exhibit significant quantum mechanical effects, such as carbon nanotubes, two-dimensional semiconductors, and recently discovered topological materials. The aim is to create new technologies for highly efficient use of solar light/thermal energy that will contribute to the realization of a sustainable energy society. To understand the unique physical properties of these materials from the fundamental principles and extract superior functions that exceed the limits of conventional materials, we are promoting interdisciplinary research that covers basic sciences, including materials synthesis and condensed matter physics, as well as thermal, mechanical, electronic, and optical engineering along with the fabrication of integrated nanomaterials. Followings are main research achievements in the year of 2023.

2. Birefringent optical responses of single-chirality carbon nanotube membranes

Membranes consisting of single-walled carbon nanotubes (SWCNTs) with a specific chirality have great potential in photonic and thermo-optic applications because of their strong and distinct light-matter interaction via excitons with high-temperature robustness. Generally, these membranes are fabricated via the vacuum filtration of single-chirality SWCNT dispersion, and SWCNTs are aligned two-dimensionally in the plane (Figure 1(a)). The electric field component normal to the membrane surface (red arrow in Figure 1(b)) is always perpendicular to the tube axis of each SWCNT. Meanwhile, the optical responses of each individual SWCNT are anisotropic with respect to the tube axis as shown in Figure 1(c). For these reasons, it is expected that the macroscopic responses of the membranes depend on the light-propagation direction. However, their anisotropic optical features and complete complex refractive index spectra remain to be unveiled, hindering the design of devices to deal with light propagating in arbitrary directions using SWCNT membranes as opto-functional materials.

In this study, we revealed the birefringent optical response of a single-chirality SWCNT membrane, as examined using polarization- and angle-resolved reflection spectroscopy [1]. We determined the ordinary (in-plane, $\tilde{n}_{in}$) and extraordinary (out-of-plane, $\tilde{n}_{out}$) complex

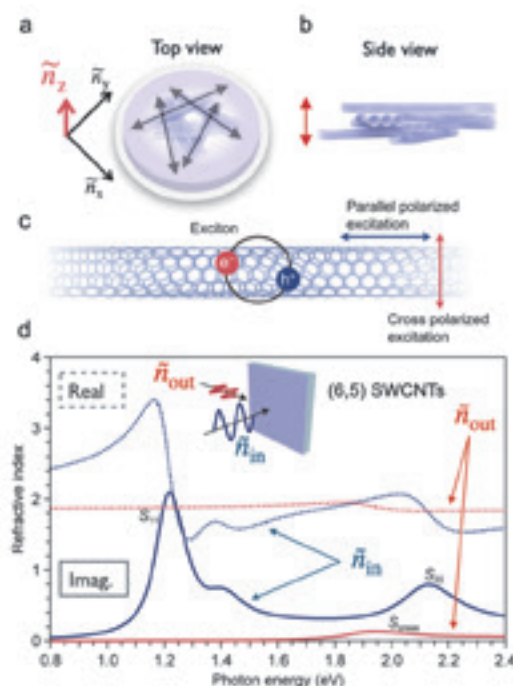


Fig. 1 Schematic of the (a) top and (b) side views of a single-chirality SWCNT membrane fabricated via vacuum filtration. (c) Anisotropy in the exciton optical response in an individual SWCNT. (d) Ordinary (in-plane, $\tilde{n}_{in}$, blue) and extraordinary (out-of-plane, $\tilde{n}_{out}$, red) complex refractive index spectra of the membrane.

refractive index spectra for it. The membrane was fabricated via the vacuum filtration of a (6,5) SWCNT dispersion prepared using gel chromatography, and the spectra were obtained in the near-infrared-to-visible region using homemade optical setups. All spectral features were consistent with a uniaxial birefringent membrane, reflecting the random in-plane orientations of SWCNTs. Figure 1(d) shows the obtained complex refractive index spectra. The ordinary one presented a series of sharp resonances of parallel-polarized excitons while the extraordinary one is ~ 1.9 with the small contribution from the cross-polarized exciton resonance (S_{cross}). We found that this resonance is small but indispensable for properly predicting angle-dependent optical responses of the membrane. The complete knowledge of the birefringence and complex refractive index spectra of the single-chirality SWCNT membrane facilitates the precise and diverse design of photonic and thermo-optic devices based on SWCNTs.

3. Fabrication of pure carbon nanotube membrane without mid- and far-infrared optical absorption

In this study, high purity single-chirality SWCNT thin films were fabricated using a polymer dispersion method, and their complex refractive index spectra were determined [2]. It was found that the Drude-type mid- and far-infrared absorption, which normally increases at lower photon energies (long wavelengths), hardly appears in the absorption spectra of high-purity semiconducting SWCNT thin films. This may be attributed to extremely suppressed thermal free carrier generation because thermally excited electrons and holes attract each other by Coulomb force to form strongly bound excitons. This study confirmed that it is possible to fabricate SWCNT thin films with almost no mid- and far-infrared absorption which causes unwanted radiative energy leak in photon-energy selective solar absorber and/or thermal emitters for efficient solar and thermal energy harvesting technologies.

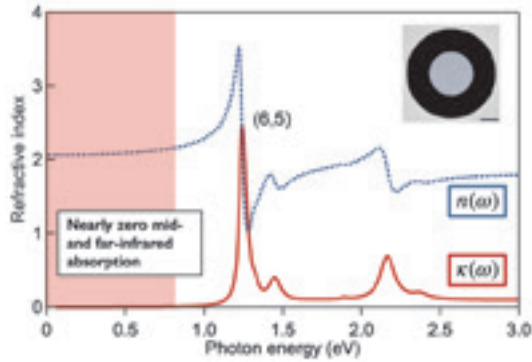


Fig. 2 Real $n(\omega)$ and imaginary $\kappa(\omega)$ parts of complex refractive index spectra of the polymer-dispersed (6,5) SWCNT film shown as the inset.

4. Exciton energy transfer under low temperature in a lateral heteromonolayer of WSe₂–MoSe₂

Controlling exciton motion in monolayer transition metal dichalcogenides (TMDCs) has been attracting much attention, because it enables new-type optoelectronic devices such as exciton transistors and exciton routers which exhibit an extremely reduced energy consumption and fast operating speed. In our previous study, the unidirectional exciton energy transport has been demonstrated in a lateral TMDC heteromonolayer at room temperature [3]. However, physical parameters related to exciton motion such as mobility and lifetime strongly depend on temperature, and it is still elusive that how temperature affects exciton kinetic property, energy transfer process.

In this study, we examined the exciton energy transfer process in a lateral heteromonolayer of WSe₂–MoSe₂ at low temperature [4]. Position-dependent photoluminescence excitation (PLE) spectroscopy measurements revealed the occurrence of exciton

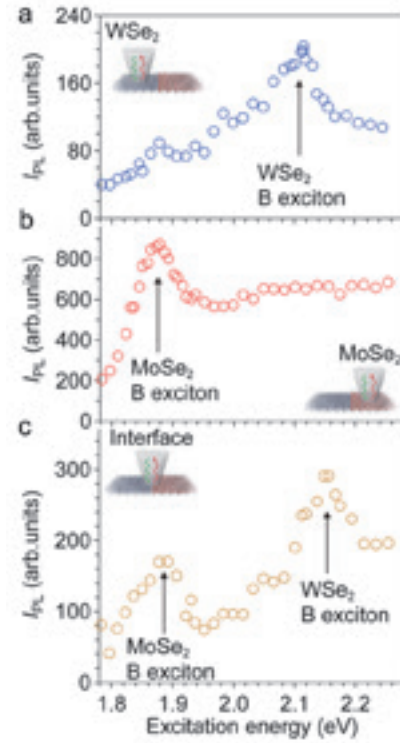


Fig. 3 Photoluminescence excitation (PLE) spectra at 15 K recorded at (a) the WSe₂, (b) the MoSe₂ and (c) the interface. The inset figures display the excitation and detection positions.

energy transport from WSe₂ to MoSe₂ both at room temperature and 15 K. The effective energy transport distance in WSe₂ was longer at 15 K than at room temperature, suggesting that the dark excitons with longer diffusion length than bright excitons preferentially contributed to the exciton energy transport across the heterojunction interface at 15 K. This study provides useful insights for the development of excitonic devices based on exciton transport in transition metal dichalcogenides.

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References

- [1] H. Wu, T. Nishihara, A. Takakura, K. Matsuda, T. Tanaka, H. Kataura, and Y. Miyauchi, *Carbon*, **2024**, 218, 118720.
- [2] T. Nishihara, Z. Liu, Y. Miyauchi, *J. Heat Transfer Soc. Jpn.*, **2023**, 62, 36.
- [3] M. Shimasaki, T. Nishihara, K. Matsuda, T. Endo, Y. Takaguchi, Z. Liu, Y. Miyata, and Y. Miyauchi, *ACS Nano*, **2022**, 16, 5, 8205.
- [4] M. Shimasaki, T. Endo, K. Watanabe, T. Taniguchi, K. Matsuda, T. Nishihara, Y. Miyata, and Y. Miyauchi, *Jpn. J. Appl. Phys.*, **2023** 62, 112002.

Collaboration Works

宮内雄平, 浙江大学 (中華人民共和国), 高純度半導体型カーボンナノチューブ試料の作製とヘテロ構造化

Financial Support

1. Grant-in-Aid for Scientific Research

宮内雄平, 挑戦的研究 (開拓), 量子非平衡吸収体を用いた太陽光熱利用の原理的革新

西原大志, 基盤研究(B), 熱励起子ポラリトン状態の実現と熱放射制御原理の開拓

川上未央子, 特別研究員奨励費, 精密熱ふく射スペクトル制御に向けた異種ナノ物質薄膜の多重積層技術の開拓

Wu Hengkai, 特別研究員奨励費, カーボンナノチューブの量子熱光物性に基づく超高効率太陽光選択吸収体の実現

2. Others

宮内雄平, 科学技術振興機構 戦略的創造研究推進事業チーム型研究 (CREST), ナノ物質科学を基盤とするサーモエレクトロニクス創成

宮内雄平, 日本学術振興会, 国際共同研究教育パートナーシッププログラム (PIRE プログラム), 配向制御ナノチューブを基盤にした日米共同クリーンエネルギー技術開発

宮内雄平, (公財) 関西エネルギー・リサイクル科学研究振興財団, 第 14 回エネルギー理工学研究所国際シンポジウムの開催

西原大志, 科学技術振興機構 創発的研究支援, ナノシステム制御による太陽光利用の技術革新

Publications

M. Shimasaki, T. Endo, K. Watanabe, T. Taniguchi, K. Matsuda, T. Nishihara, Y. Miyata, Y. Miyauchi, Exciton energy transfer under low temperature in a lateral heteromonomolayer of WSe₂-MoSe₂, Japanese Journal of Applied Physics, 62, 11, 112002, 2023

H. Wu, T. Nishihara, A. Takakura, K. Matsuda, T. Tanaka, H. Kataura, Y. Miyauchi, Birefringent optical responses of single-chirality carbon nanotube membranes, Carbon, 218, 118720, 2023

西原大志, 劉知銳, 宮内雄平, 中遠赤外光吸収を持たない純炭素薄膜~カーボンナノチューブの熱ふく射制御応用に向けて~, 伝熱, 62, 260, 36-42, 2023

西原大志, 自作簡易エリプソ分光による熱ふく射物性計測, 熱物性, 37, 3, 100-104, 2023

Presentations

T. Nishihara, Nanophotonics relying on distinct thermo-optical properties of low-dimensional excitons, ICNN2023(International Conference on Nano-photonics and Nano-optoelectronics), パシフィコ横浜, 2023.4.19

Y. Miyauchi, Photophysics and chemistry of carbon nanotubes at high temperatures, 2023 Workshop on JUNCTIONS: Japan-US Network for Clean energy Technologies Involving Oriented Nanotubes, Jyobun Hall, Yokohama Media and Communications Center, 2023.5.22

Y. Miyauchi, Anisotropy in complex refractive index spectra of single-chirality-enriched carbon nanotube films, The 243rd ECS Meeting, 2023, Hynes Convention Center, Boston, 2023.5.30

西原大志, Photon's free energy classifying one-dimensional excitonic light emission, 第 17 回光物質科学研究討論会, 京都大学, 2023.6.3

西原大志, 自作簡易エリプソ分光による熱ふく射物性計測, 日本熱物性学会 セミナーシリーズ, オンライン, 2023.6.30

西原大志, ナノ物質のユニークなエネルギー変換: 熱から単一波長の光へ, 第 46 回京都大学宇治キャンパス産学交流会, 京都大学宇治キャンパス, 2023.7.5

Y. Miyauchi, Studies on optical properties of single-chirality carbon nanotube membranes for photothermal applications, The Tenth US-Japan Joint Seminar on Nanoscale Transport Phenomena, Loews Coronado Bay Resort, San Diego, California, USA, 2023.7.17

西原大志, アップコンバージョン分光器を用いたロックイン”ライクな”中赤外分光, ふく射勉強会, 東京工業大学, 2023.8.26

Y. Miyauchi, Study on high-purity carbon nanotube membranes toward solar energy harvesting and utilization, The 14th International Symposium of Advanced Energy Science, 京都大学宇治キャンパス, 2023.9.1

西原大志, 放射エントロピーで再定義する多彩な 1 次元励起子発光, 日本物理学会第 78 回年次大会, 東北大学, 2023.9.18

H. Wu, T. Nishihara, A. Takakura, K. Matsuda, T. Tanaka, H. Kataura, Y. Miyauchi, Determination of in- and out-of-plane complex refractive index spectra of single-chirality carbon nanotube membranes, 第 84 回応用物理学会秋季学術講演会, 熊本城ホール, 2023.9.20

宮内雄平, カーボンナノチューブの量子物性科学に基づく太陽エネルギー利活用の新たな可能性, 第 3 回ナノカーボン未来技術講演会「カーボンニュートラルを実現するナノカーボンの未来技術」, 東京大学弥生講堂一条ホール, 2023.10.20

西原 大志, 低次元励起子の特異な熱光物性と高温ナノフォトニクスへの応用, 第 8 回フォトニクスワークショップ「光で照らす多彩な世界」, 沖縄県青年会館, 2023.11.2

Y. Miyauchi, Science and engineering of excitons in low-dimensional nanomaterials, 36th International Microprocesses and Nanotechnology Conference (MNC 2023), Keio Plaza Hotel Sapporo, Hokkaido, Japan, 2023.11.15

T. Nishihara, Distinctive high-temperature light emission originating from one-dimensional excitons of carbon nanotubes, Workshop on Innovative Nanoscale Devices and Systems 2023, Outrigger Kona Resort and Spa, USA, 2023.12.5

宮内雄平, 熱と光とナノ物質の科学, 第 1 回ナノシステム研究会, 島津製作所本社, 2023.12.2

宮内雄平, ナノ物質の励起子光物性と熱放射研究, 領域全体会議, TKP カンファレンスセンター, 2024.2.17