

## 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

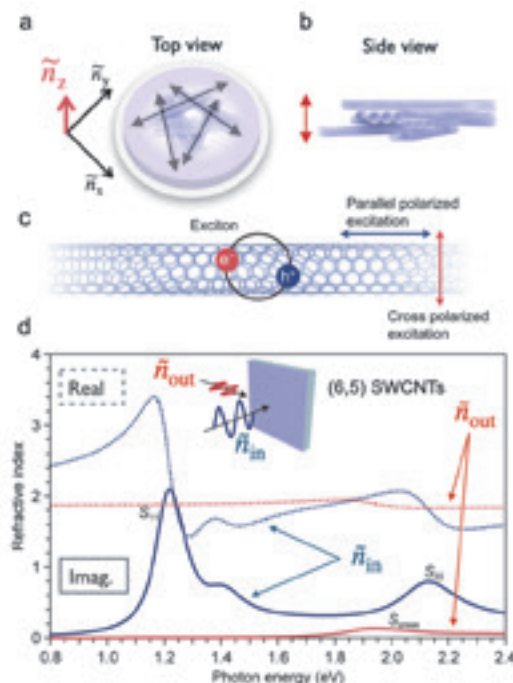


Fig. 1 Schematic of the (a) top and (b) side views of a single-chirality SWCNT membrane fabricated via vacuum filtration. (c) Anisotropy in the exciton optical response in an individual SWCNT. (d) Ordinary (in-plane,  $\tilde{n}_{in}$ , blue) and extraordinary (out-of-plane,  $\tilde{n}_{out}$ , red) 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  $\sim 1.9$  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.

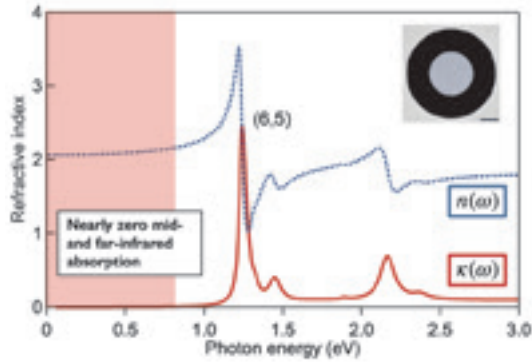


Fig. 2 Real  $n(\omega)$  and imaginary  $\kappa(\omega)$  parts of complex refractive index spectra of the polymer-dispersed (6,5) SWCNT film shown as the inset.

### 4. Exciton energy transfer under low temperature in a lateral heteromonolayer of WSe<sub>2</sub>–MoSe<sub>2</sub>

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 WSe<sub>2</sub>–MoSe<sub>2</sub> at low temperature [4]. Position-dependent photoluminescence excitation (PLE) spectroscopy measurements revealed the occurrence of exciton

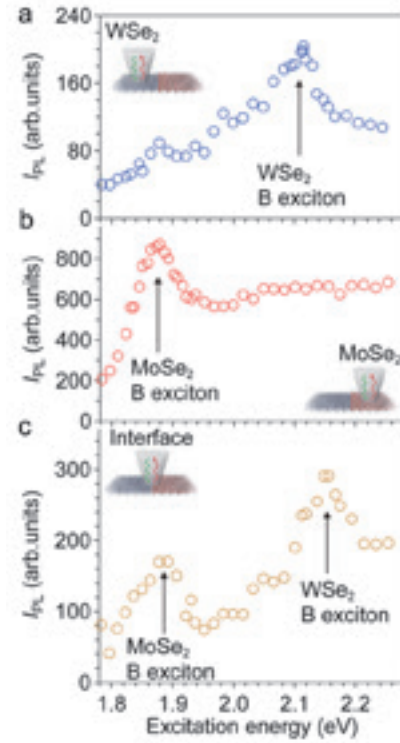


Fig. 3 Photoluminescence excitation (PLE) spectra at 15 K recorded at (a) the WSe<sub>2</sub>, (b) the MoSe<sub>2</sub> and (c) the interface. The inset figures display the excitation and detection positions.

energy transport from WSe<sub>2</sub> to MoSe<sub>2</sub> both at room temperature and 15 K. The effective energy transport distance in WSe<sub>2</sub> 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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## 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

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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 WSe<sub>2</sub>-MoSe<sub>2</sub>, Japanese Journal of Applied Physics, 62, 11, 112002, 2023

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