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 (CNTs), two-dimensional (2D) 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 2022.

2. Statistical verification of anomaly in chiral angle distribution of air-suspended carbon nanotubes

CNTs have long attracted attention due to their distinct optical, mechanical, electronic, and thermal properties, which are beneficial in the development of various future technologies. In addition, their potential to be generated from carbon dioxide has extended their importance in terms of present social demands to reduce carbon dioxide emissions. Among CNTs, single-walled CNTs (SWCNTs) have excellent functionalities that are sensitively dependent on their chiral structures, which are determined according to their diameter and chiral angle. However, large-scale bulk synthesis of SWCNTs with an on-demand chiral structure is still difficult and clarifying their growth mechanism in detail toward structure-controlled bulk synthesis is of central importance in nanomaterials fields. To date, theoretical studies have predicted that the configurations of the opening tube edge composed of armchair- and zigzag-shaped sites have significant impact on the SWCNTs' growth rate and the resultant structure abundance distribution. However, lack of sufficient statistical data on the chiral structure distribution of both metallic and semiconducting SWCNTs has prohibited rigorous experimental verification of such theoretical predictions.

In this study, we reported statistical verification of the chiral structure distribution of as-grown, air-suspended individual SWCNTs that are unaffected by various nonideal effects, including interactions with other nanotubes in bundles and with the substrate [1]. We determined the chiral structures of 413 air-suspended SWCNTs using broadband Rayleigh spectroscopy. Our statistical analyses have revealed that the fractional abundance as a function of chiral angle shows a linear increasing trend with an anomaly in the derivative at the chiral angle of approximately 20° (refer to Fig. 1). From this critical result, we have clarified that the consecutive armchair site (red edge in the inset) is a key edge configuration strongly influencing the growth preference of each SWCNT structure. These findings on the anomalous abundance distribution and the proposed model accounting for the chiral angle dependence of the growth rate are expected to allow us to obtain deeper understanding of the growth mechanism of SWCNTs.

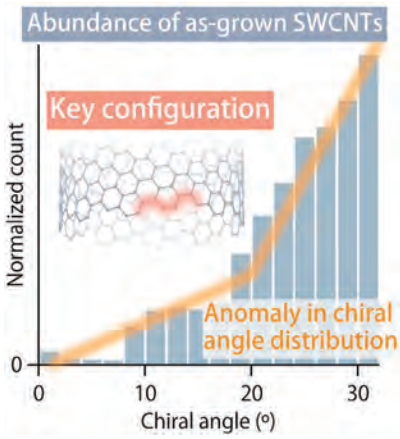


Fig. 1 Normalized chiral angle distribution of as-grown, air-suspended individual single-walled carbon nanotubes (SWCNTs) [1].

3. Directional exciton-energy transport in a lateral heteromonolayer of WSe_2–$MoSe_2$ (Fig. 2)

Excitons, in particular those in low-dimensional semiconducting nanomaterials are expected as next generation energy and/or information carriers for future energy/information devices. For these devices, controlling of the direction of exciton energy flow is one of the most important challenges. Generally, spatial asymmetry is required to induce a directional flow of quasi particles. As for charged particles like electrons, generation of such a flow is simply achieved by introducing potential asymmetry using an external electric field. However, for generating a directional

flow of neutral particles such as excitons, electric field cannot be a direct driving force. Therefore, finding a method to directionally drive excitons laterally in an ultrafast time scale has been strongly desired.

We reported observation of directional exciton energy transport across the interface in the WSe_2-$MoSe_2$ lateral heterostructure [2]. Space and frequency resolved photoluminescence (PL) and PL excitation measurements revealed exciton energy transfer in the lateral direction induced by asymmetric energy landscape bult-in at the sharp interface of the two different 2D semiconductors. The results suggest that excitonic devices using this type of lateral heterostructure is promising in applications for energy/information devices using fast exciton energy transport.

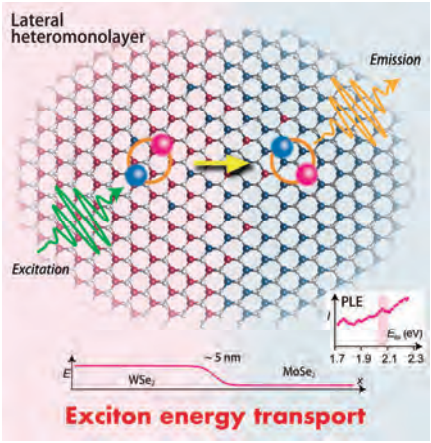


Fig. 2 Directional exciton-energy transport in a lateral heteromonolayer of WSe_2–$MoSe_2$ [2].

4. Anisotropic exciton drift-diffusion in a monolayer WS_2xSe_{2-2x} alloy with a gradually changing composition

In addition to the demonstration of the unidirectional transport of excitons using lateral heterostructure monolayer with a sharp one-dimensional heterojunction [2], we also demonstrated anisotropic exciton drift-diffusion in a $WS_{2x}Se_{(2-2x)}$ alloy monolayer with gradually changing composition ratio extended over micrometers [3]. The significant observation was that the spatial profile of the PL image became anisotropic under the finite exciton energy gradient under isotropic laser excitation. We confirmed that the degree of asymmetry had a strong correlation with the energy gradient. The results were successfully interpreted based on the simulation results considering drift and diffusion of excitons under the finite exciton energy gradient.

5. Polarized Raman spectroscopy on topological semimetal $Co_3Sn_2S_2$

In the last few years, the ternary ferromagnetic compound $Co_3Sn_2S_2$ has been identified as a topological semimetal, which can host a Weyl semimetal phase. To date, electronic properties of this material has attracted growing attention because it provides a desirable platform to study Weyl fermion physics near the Weyl point that acts as a source or sink of the Berry curvature field, which is one of the central interests of the current condensed matter physics. Moreover, Weyl semimetals is expected as a candidate material for nonreciprocal thermal emitters that break Kirchhoff's law, which may lead to a breakthrough in the radiative heat transfer technologies. In contrast to the intensively studied electronic properties, however, phonon properties of $Co_3Sn_2S_2$ have yet to be clarified experimentally; even the most fundamental information, phonon frequencies and their symmetries have unidentified. The lack of the Raman spectral fingerprint in the literature has hindered a quick experimental identification of the crystal structure, as well as confirmation of the validity of ab-initio calculations on $Co_3Sn_2S_2$ by comparing the calculated phonon frequencies with the experimental ones.

In this study, we clarified polarized Raman spectra of the topological semimetal $Co_3Sn_2S_2$ [4]. Two major phonon Raman peaks were observed at 289 cm^{-1} and 386 cm^{-1} over continuous background emission signals, and attributed to Γ point phonon modes with E_g and A_{1g} symmetries, respectively, according to the group theory and Raman tensor analysis. Line shape analyses revealed that the high-frequency A_{1g} mode exhibited asymmetric peak feature suggesting the Fano resonance between the A_{1g} phonon scattering with the continuous electronic background. The clarified phonon energies and symmetries, as well as the electronic contribution to the Raman scattering, will not only be useful as a fingerprint to readily verify the experimentally grown or theoretically calculated crystal structure, but also suggest importance of Raman spectroscopy as an effective tool to study low energy excitations and their interactions in $Co_3Sn_2S_2$.

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