Abstract

Study on photoluminescence quantum yields of atomically thin-layered two-dimensional semiconductors transition metal dichalcogenides

by

Nur Baizura Binti Mohamed, Ph.D.

Kyoto University, Japan (2018)

Advisor: Professor Dr. Kazunari Matsuda

In order to confront with the natural energy shortage in the world, the intensive efforts have been forced to fulfill the demands on the technological applications such as low energy consumption electronic and optical devices. The emergent new materials and their technological applications are strongly required to fulfil these demands. Atomically thin-layered two-dimensional (2D) materials have been attracted and stimulated the research interests due to their intriguing electronic and optical properties, after the discovery of an atomically thin carbon material of graphene in 2004. The semiconducting monolayer transition metal dichalcogenides (TMDs), MX_2 (M = Mo, W, Re; X = S, Se) with superior and divergence electronical and optical properties, would have high potential for these requirements, while the graphene without a band gap is limited towards the optoelectronic devices applications.

The optical properties of atomically thin TMDs have dominated by bound electron-hole pairs (excitons) by Coulomb interactions, as a consequence of extremely large exciton binding energy of several hundred meV. The large binding energy comes from strong quantum confinement of electron and hole in the atomically thin space and reduced dielectric screening in the atomically thin materials, and this large binding energy enables the stable excitons even at room temperature. The exciton related optical properties are very crucial to be investigated for understanding the light-matter interaction, for instance, the light emission

quantum yield (QY) and radiative lifetimes of elementary excitation, in the atomically thin layered materials.

In this thesis, I have developed the method to evaluate the photoluminescence (PL) QY of atomically thin-layered material with combination of absolute and relative methods. The important characteristics in choosing the reference dye is that the assigned dye should have high and stable QY value in a solid phase, as well as show almost same emission wavelength with those in TMDs materials. The PL QY value is a very crucial parameter to determine the light emission efficiency and the performance of light-emitting devices. Moreover, the PL QY is also a sensitive probe to identify the electronic structure of materials. I have applied the developed PL QY method to identify the nature of electronic structure of rhenium disulfide (ReS_2), which has not been fully understood and debated to-date. Then, I extended the study to reveal the intrinsic radiative lifetime of excitons of monolayer MX_2 (M = Mo, W; X = S, Se) by combining the PL QY and PL decay measurements. The long effective radiative lifetime of excitons in monolayer MX_2 at room temperature could be understood in the basis of three factors: (1) long radiative lifetime at low temperature limit, (2) finite coherence area of excitons with several square nanometers, and (3) populations of dark exciton states.

The development of this PL QY method of atomically thin-layered material can be significant for the evaluation of new thin-material with unrevealed electronic structure. Moreover, the obtained results of intrinsic physical value of monolayer TMDs are expected to contribute for understanding the physics of light-matter interaction and application of atomically thin-layered materials, to overcome the challenges toward the novel atomically thin optoelectronic devices for future next generations.