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Monochromated EELS to Probe the Local Optical Properties of Low-Dimensional Materials

Kazu Suenaga[1], Luiz H. G. Tizei[1,2], Yung-Chang Lin[1] and Ryosuke Senga[1]

[1.] National Institute of Advanced Industrial Science and Technology (AIST), Central 5, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan
[2.] Laboratoire de Physique des Solides, Universite Paris-Sud, Bat 510, Orsay 91405, France

In this presentation, we show our recent studies of low-dimensional materials by means of high-energy resolution EELS. In the scheme of the triple C project we have been developing low-voltage TEM/STEMs working at low acceleration voltages ranging from 15 to 60 kV. One of the microscopes has recently mounted a double Wien-filter type monochromator which is placed before the acceleration tube in a schottkey gun [1]. A largest advantage of this system is the simple use to achieve the desired energy width just by changing the slit size on the fixed dispersion plane without further re-adjustment of the microscope alignments (Fig. 1). This system has already proved excellent performances in the TEM mode when equipped with the higher-order aberration corrector (the DELTA model) [2]. A low-voltage GIF quantum is installed and quite useful for acquiring sequential spectra in the spectrum-image mode.

When used in the STEM-EELS mode, high-energy resolution can be achievable as 200 meV in FWHM for core-loss measurement and 30 meV for low-loss with the atomic resolution retained. It though sacrifices the probe current substantially as seen in the Fig. 1. A highest energy width available ~14meV is therefore unpractical to use. The conventional side-entry specimen holders allow us experiments in the various temperature ranges from -180C to 550C. Acquiring more than hundreds of spectra at different probe position indeed requires the dual EELS mode simultaneously recording the unsaturated zero-loss peak that can assure a post-processing for the precise energy calibration. Spectral defocus on the core-loss due to the chromatic aberration of post-specimen lens is also an important issue and must be corrected *in situ*.

Fig. 2 shows an example of spatially resolved exciton mapping across an interface between single-layered MoS_2 and $MoSe_2$ [3]. The experiment was performed at -120C with a few nanometer probe at 30kV. The spatial resolution of EELS mapping is supposed to be determined by the size of exciton. Atomic resolution imaging is needed to confirm the desired structure and to avoid the contaminated/defective areas. Two distinct absorption spectra can be obtained across the interface. The change from MoS_2 to $MoSe_2$ is followed by peak broadening but no apparent spectral shift is found. No interface mode has been found so far. Cryo-stage is indispensable to measure the sharp exciton peaks in a series of transition metal dichalcogenide materials.

References

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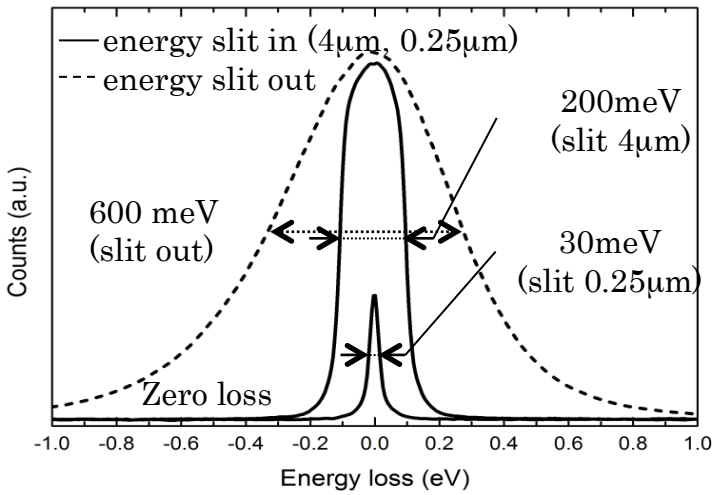

Figure 1. Zero-loss profiles when the monochromator is excited (Senga et al., [4]). Energy resolutions of 600 meV (without the slit), 200 meV (4-μm slit inserted), and 30 meV (0.25-mm slit inserted), was achieved and measured by the zero-loss peak at the FHWM. Each peak area corresponds to the relative probe current.

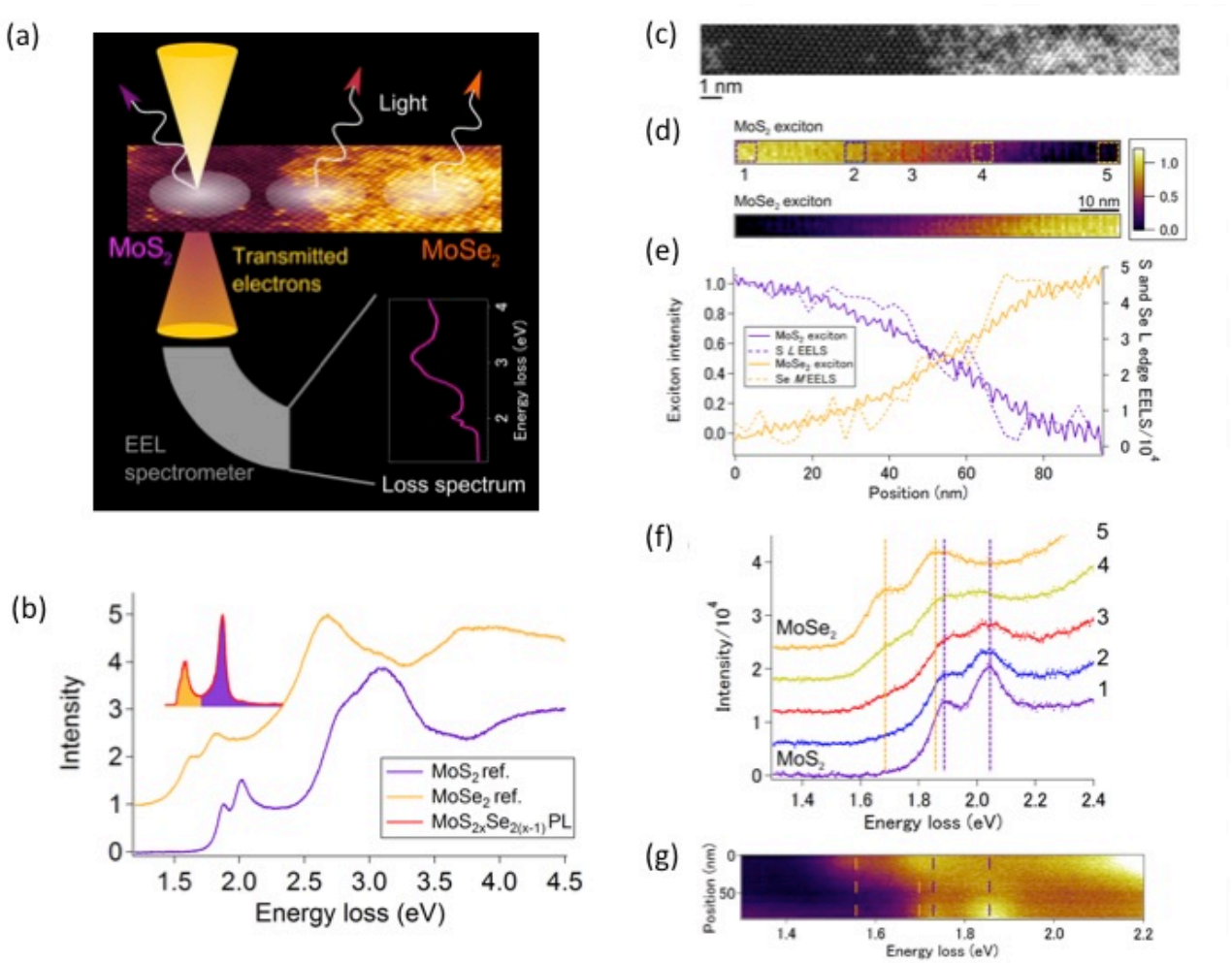

Figure 2. STEM-EELS on a heterojunction of MoS_2-$MoSe_2$ (Tizei et al. [3]). (a) EELS in transmission across an interface. (b) PL peak and two reference spectra. (c) ADF image of a diffuse interface. (d) (e) Fitting coefficients for MoS_2 (above) and $MoSe_2$ (below) and the chemical profiles. (f) Spectra integrated at different positions across the interface marked in (d). (g) Spectrum-line in the spatial direction perpendicular to the interface. Specimen courtesy of Prof. Li in KAUST.

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