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Nanoscale optical and vibrational spectroscopy of low-dimensional materials

Physical properties of low-dimensional materials are strongly influenced by the quasiparticle excitations at non-perfect structures like boundaries, edges, defects or irregular stacking sequence. Unfortunately, such local information has been usually averaged in conventional inelastic scattering techniques using x-ray, neutron and light sources because of their inferior spatial resolution. Here we demonstrate the nanoscale optical and vibrational spectroscopy of 1D/2D materials by using a monochromatic electron source mounted in a scanning transmission electron microscope. Its energy resolution, better than 30 meV, allows to access the quasiparticle excitations (i.e. phonon, exciton and plasmon) of low-dimensional materials by electron energy-loss spectroscopy (EELS). The spatial and momentum resolutions are dependent on each other and can be tuned freely. For instance, by integrating a wide momentum space, an atomic sized probe can be formed and the local spectroscopy on a single defect is possible. Indeed, we have successfully measured the optical gap transitions from a defect of an individual semiconducting carbon nanotube [1,2]. The optical conductivity extracted from an EEL spectrum via Kramers-Kronig relation for a certain type of defect presents a characteristic modification near the first exciton peak (Fig. 1). The line-width of exciton peak shows a variety of broadening at different defect sites and suggests different degrees of shortening of its lifetime. In contrast, an electron probe with a higher momentum resolution can provide a full phonon dispersion of 2D materials such as hexagonal boron nitride or graphene at a few tens nanometre scale. This local spectroscopy with a large flexibility will open up a wide possibility to unravel the defect physics of quantum matters.

References

[1] R. Senga, T. Pichler and K. Suenaga, Nano Letters. 16, (2016) 3661.
[2] R. Senga, T. Pichler, Y. Yomogida, T. Tanaka, H. Kataura and K. Suenaga, Nano Letters. 18, (2018) 3920.

Figures

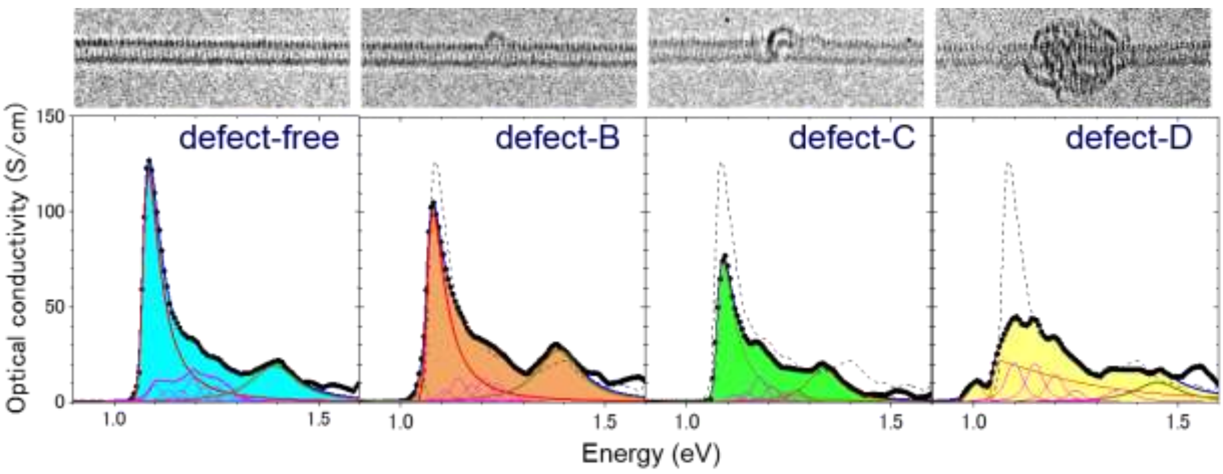

Figure 1: Absolute optical conductivity of a semiconducting (9,2) at three different defects as well as a defect-free region. This shows how the gap transition and exciton lifetime depend on the type of local defect [2].