Ultrafast Photo-Response of MOF Based Quantum Magnet

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Metal-organic framework (MOF) material, $(NPr_4)_2[Fe_2(Cl_2An)_3]$, which can be classified as a one-dimensional magnet in the low-temperature phase, has been confirmed to show the two-step charge-transfer (CT) phase transition (T_c = 317 and 354 K) between Fe and Cl_2An ions based on magnetic, structural, and spectroscopic measurements [1]. The structural changes at T_c are minimal due to relatively strong chemical bonds among constituents. Then the phase transition can be viewed as a purely electronic one. Therefore, the ultrafast and highly efficient CT phase transition can be expected by photoexcitation. Indeed, we confirmed the highly efficient CT transition occurs within 100 fs by time-resolved spectroscopy exciting CT transition (~0.8 eV) and probing the near-IR to UV energy range at room temperature (RT) [2,3]. We extended our study into the mid-IR (molecular vibrational) range to reveal the structural changes accompanied by this CT phase transition.

As shown in Figure 1, photoinduced CT phase change indeed occurs just after the photoexcitation of 0.8 eV. However, the observed spectral shape is different from that observed in any thermally induced phases, significantly below about 0.25 eV. The experimental results strongly suggest the photoinduced appearance of electron-molecular vibration coupled (e-mv) mode (the appearance of the dynamically dimerized state which never appears by thermal excitation (hidden state). The precise dynamics will be discussed in the presentation. The development of an electron diffraction system for observing structural dynamics with sub 100 fs time-resolution in MOF and related materials will also be addressed.

References:

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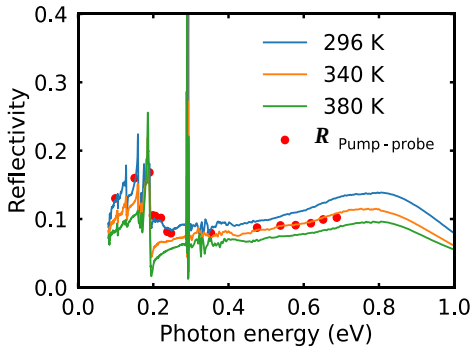

Figure 1: The temperature-dependent reflectivity spectra with the polarization perpendicular to the c-axis (solid lines), and the transient reflectivity spectrum immediately after photoexcitation (red circles). Sharp structures at around 0.28 eV are due to the CO_2 absorption.