

## Realizing Domain Excitation with High-Energy Photons: Inelastic X-Ray Scattering and Angle-Resolved Photoemission

We describe a process in which a certain block of electrons in a particular phase in a solid are excited simultaneously by a single high-energy photon and converted into a domain of a different phase. Based on energetics, we expect this phenomenon to occur at least in the vicinity of the phase boundary, and numerically reliable calculations confirm this for a one-dimensional photo-induced phase transition system. As possible experiments to observe the phenomenon, we propose inelastic X-ray scattering (IXS) and angle-resolved photoemissions (ARPES). In particular, ARPES can provide rich information associated with the spin degrees of freedom.

In the conventional picture of photo-excitation, a single photon excites only one electron. This is true for many known systems of solids, to which one-electron pictures are successfully applied. On the other hand, there are completely different situations where photo-excitation is always accompanied by the excitation of multiple electrons. One such situation is idealistically realized in photo-induced phase transition systems [1]. Figure 1 illustrates the case of a quasi-one-dimensional neutral-ionic transition system having donors (D) and acceptors (A) alternatively aligned. After IXS, we first expect an exciton-like state as depicted in Fig. 1(a). In contrast to cases in ordinary insulators, the excitation does not stop at this state but proliferates through domain states such as those shown in Figs. 1(b) and (c) and so on. The driving force of this proliferation is the intrinsic electron transfer (to), combined with the energetic degeneracy among the states.

Before going into further details, we explain what it means to detect a domain via spectroscopy. In a sense, a domain is a spatially extended particle which can

propagate in a crystal. This simple statement means that the domain has at least two fundamental degrees of freedom -- its center of gravity and its spatial size. As is the case for ordinary particles, the former leads to energy dispersion for finite momenta. Regarding the latter, we point out that domain sizes are generally quantized except exactly at phase boundaries. On the whole, we expect spectra that consist of many size-related branches with finite dispersions. What we emphasize here is that for both IXS and ARPES, which we discuss later, we only need a single excitation. Namely, this is not a so-called pump-probe experiment that requires much more complicated apparatus.

Our calculated IXS spectra have confirmed the above concept [2]. Since the details are already contained in a highlight article of Activity Report 2006, we do not repeat them here and just mention future prospects. Currently, we are planning to detect such effects in actual materials. The most promising material is an organic molecular solid, TTF-CA, which is known to exhibit an ultra-fast photo-induced phase transition on

### Inelastic X-ray scattering

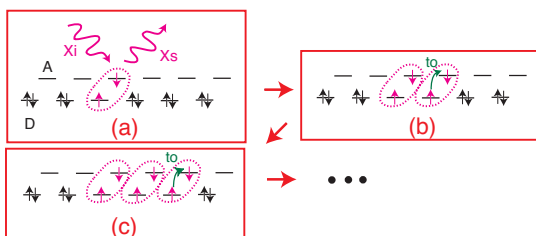


Figure 1

Schematic illustration for the proliferation of a domain in the neutral-phase background following inelastic X-ray scattering. The singlet spin pairs are surrounded by dotted lines. In each level diagram, the lower (upper) levels are those for the HOMO (LUMO) of the donor (acceptor) molecule. "D" and "A" correspond to a donor and an acceptor, respectively.

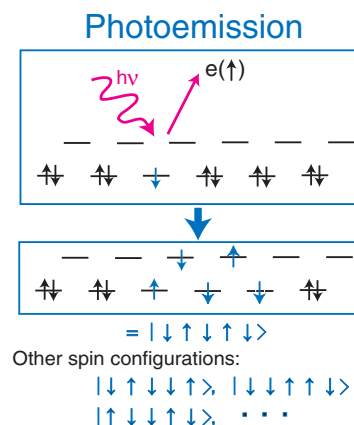


Figure 2

Same as Fig. 1, but for the case of photoemission. The possible spin configurations inside a domain are expressed as  $|\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\rangle$ .

a time scale shorter than 10 fs. In IXS experiments on TTF-CA, it will be important to utilize resonant effects related to the S (sulfur) absorption edge ( $\sim 2.4$  KeV), since the weights of the S orbitals are large in the molecular orbitals around the energy gap. This stimulates our further efforts both in theory and in experiment, in the sense that the resonance condition must be included in our calculation and that new soft X-ray IXS equipment targeting the S absorption edge must be developed.

Photoemission also gives rise to domain formations triggered by one-electron removal, instead of the exciton excitation [2]. When we watch this process carefully, we encounter much more entanglement among the degrees of freedom. In particular, the spin degrees of freedom combine with the other degrees of freedom to strongly modify the spectra, as shown in Fig. 2.

In Fig. 3, we show calculated ARPES for two momenta ( $k$ ), at the zone center (left) and at the zone boundary (right). If we only consider the two basic degrees of freedom, the spectrum for the zone center

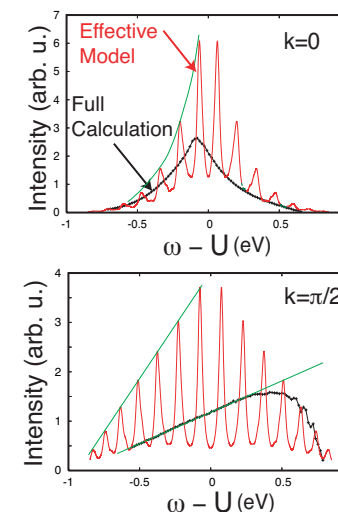


Figure 3

ARPES spectra expected at the phase boundary. The black lines indicate the results of the full calculation, the red lines those obtained by effective-model analyses. The green lines are guides for the eyes. The binding energies are measured from the on-site Coulombic repulsion energy  $U$ . For the parameter values, see Ref. 2.

should shrink to a sharp peak. However, the spectrum turns out to be very different, taking a cusp-like structure. Furthermore, the spectrum at the zone boundary also takes a spectacular shape, showing a triangle-like structure. All these features are qualitatively reproduced by an effective model that incorporates spin degrees of freedom, as plotted by the red lines in Fig. 3.

To conclude, we have demonstrated how domain excitations manifest themselves in IXS and ARPES spectra, leading to spectral variety characteristic of each measurement. We hope for future experimental studies, believing that these would deepen our basic understanding of the many-body nature of excited states in solids.

### REFERENCES

- [1] K. Iwano, *Phys. Rev. Lett.*, **97** (2006) 226404.
- [2] K. Iwano, *Phys. Rev. Lett.*, **102** (2009) 106405.

K. Iwano (KEK-PF)