Direct Domain Excitation by Photoemission and Its Dynamical Features Manifesting in ARPES spectra

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The theoretical and experimental studies of angle-resolved photoemission (ARPES) have focused on quasi-particle properties in various kinds of materials. Among them, heavily renormalized nature of quasi-particles typically seen in strongly-correlated electron systems have collected much attention, since the nature can provide important information on the many-body properties of those systems. However, even in such cases the quasi-particle basically consists of one hole; the strong renormalizaiton only increases the particle mass and the lifetime. In this sense, what we observe in ARPES is inevitably of one-hole nature, however it is modified by interactions.

In this paper, we report on a completely different type of excitation that can be detected in the ARPES. In Fig. 1, we show a schematic picture in which a system that is located close to a phase boundary absorbed one high-energy photon. Assuming a valence photoemission, we are left with one hole in the valence band. In ordinary insulators, the process stops at this point. However, in this system, it does not stop but proceeds to form a domain [1]. We here think of a one-dimensional system whose ground state is expressed as in Fig. 1(a). TTF-CA, an organic molecular solid, is a system appropriate for this situation, having nearly degenerate neutral (N) and ionic (I) phases. In the process shown in Fig. 1(b), we expect that a domain of the I phase is created in the background of the N phase. It is emphasized that the driving force is the proximity to the N-I phase boundary combined with inherent electron itineracy, and that the detection of this process requires no pump-probe experiment but only an ordinary type of photoemission measurement.

In Fig. 2, we show calculated ARPES spectra (black lines) at two momenta, which are obtained very close to the phase boundary [2]. What is quite unique is that each spectrum exhibits a special spectral shape; the spectrum at k=0 takes a cusp-like shape, while the left-hand side of the spectrum at k=π/2 is almost fitted to a straight line. We also tried deriving an effective model that retains the two basic degrees of freedom of a single domain, namely, its center of gravity and its spatial size, plus spin degrees of freedom inside the domain. As a result of its analysis, we find that the effective model reproduces the spectral features, as shown by the red lines. In particular, the above-mentioned spectra shapes are governed by the spin degrees of freedom, in a way that domain states with various spin configurations form a Hilbert space that can be accessed via the ARPES measurement.

This type of domain excitation is new in the ARPES studies, in the sense that it has never been observed so far. In addition to this, it also has important meaning in the context of photoinduced phase transitions. We hope for future experimental attempts that try to identify them in actual materials.

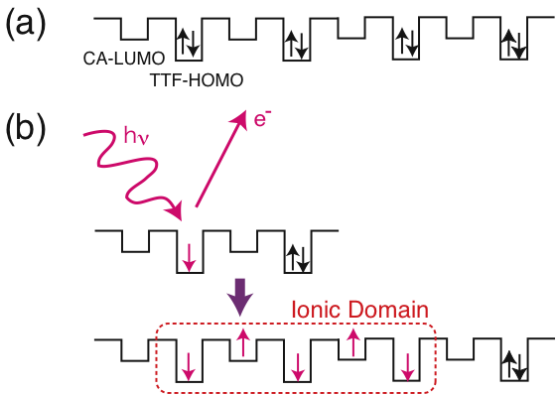

Fig. 1. (a) Neutral ground state and (b) schematic picture of an I-domain formation process.

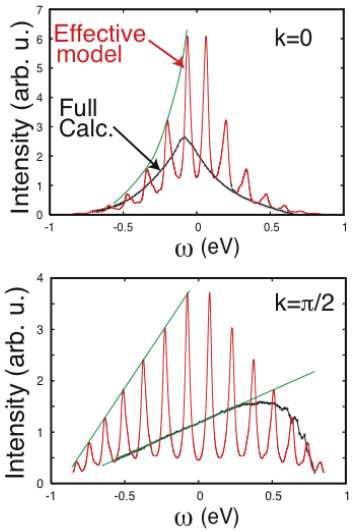

Fig. 2. Calculated ARPES spectra.

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