12 Theory

12-1 Non-Localization of Both Types of Carriers in a Quasi-One-Dimensional Strongly Correlated Electron System

The interplay between the two basic interactions in solids, i.e., electron-lattice (e-l) and electron-electron interactions, is a hot and important issue that has been continually discussed since the discovery of high-T_c superconductivity in cuprates. Although the finding of a so-called *kink* structure appearing in the dispersions of angle-resolved photoemission spectra (ARPES) [1] has without doubt stimulated many studies, it seems that its understanding is still controversial in the cuprates. In our opinion, one of the main reasons could be their rather complicated structures. Consequently, it is essential to consider both in-plane and out-of-plane oxygen movements in the cuprates, the roles of which have not yet been made clear.

In this study, we look at the same problem with a different material; quasi-one-dimensional halogen-bridged Ni compounds having the chemical formula of $[Ni(chxn)_2X]X_2$ with X representing either Br or Cl. A Mott-insulator state is realized on the central chain of the compound in which Ni^{3+} and X^{-1} ions are alternately arranged. The compound also contains a ligand chxn around each Ni^{3+} ion and counter ions X^{-1} between the chains. At first glance this system seems rather complicated. However, low-energy excitations are confined within the central NiX chain. We also expect that the e-l interactions are almost limited to the chain, thus making the problem easier to tackle than that in the cuprates.

The model Hamiltonian used here is written as

$$H = -\sum_{l\sigma} t(l)(C^{\dagger}_{l+1\sigma}C_{l\sigma} + h.c.) + \sum_l (e(l)+\delta e(l))n_l$$
$$+\sum_{l=odd} U_p n_{l\uparrow}n_{l\downarrow} + \sum_{l=even} U_d n_{l\uparrow}n_{l\downarrow}$$
$$+\sum_l V(l)n_l n_{l+1} + \sum_{l=odd} V_{pp} n_l n_{l+2} + \sum_{l=even} V_{dd} n_l n_{l+2}$$
$$+\sum_l \frac{K_l}{2} Q_l^2 \quad . \qquad (1)$$

We use a kind of *dp* model that assumes a one-dimensional array of the Ni $3d_{z^2}$ orbitals and the X p_z orbitals at the even- and odd-numbered sites, respectively, as shown in Fig. 1. Although the reader should refer to [1] for the details of eq. (1), we write here explicitly its most important part, i.e., the e-l terms in the first, second and fifth terms;

$$t(l) = t_{dp} - \beta(Q_{l+1} - Q_l) \quad , \qquad (2)$$

$$e(l) = \begin{cases} e_p - 2\alpha(Q_{l+1} - Q_{l-1}) & \text{for odd } l \\ e_d + \alpha(Q_{l+1} - Q_{l-1}) & \text{for even } l \end{cases} \quad , \qquad (3)$$

$$V(l) = V_{dp} - \alpha(Q_{l+1} - Q_l) \quad . \qquad (4)$$

The ground state in the hole picture is schematically shown in Fig. 1(a). The compound is a typical Mott insulator originating from the large on-site Coulomb energy U_d at the Ni sites. When one hole is added to or taken away from the system, one might imagine that the states will resembe those in Figs. 1(b) and 1(c), since it is "common sense" that carriers are always localized in one dimension.

Here, we disprove such "common sense", demonstrating that the carries are spatially extended in most of the Mott-insulator region of the phase diagram [2]. Figure 2 shows our main results for both the electron and hole carriers. S_1 and S_2 are the degrees of site-diagonal and site-off-diagonal e-l interactions expressed as α^2/K and β^2/K, respectively, with the dimension of energy. Here we only consider the halogen movements, setting $K_{l=even} = \infty$ and $K_{l=odd} = K$. Roughly speaking, the role S_1 plays in the localization is to cause an extra downward shift of the site energy, while S_2 makes a bond shrink so as to realize a charge-localized bond and/or an enhanced exchange coupling between two neighboring spins. Our conclusion here is that the carriers are NOT localized except for a narrow region near the phase boundary, even in the presence of the rather strong e-l

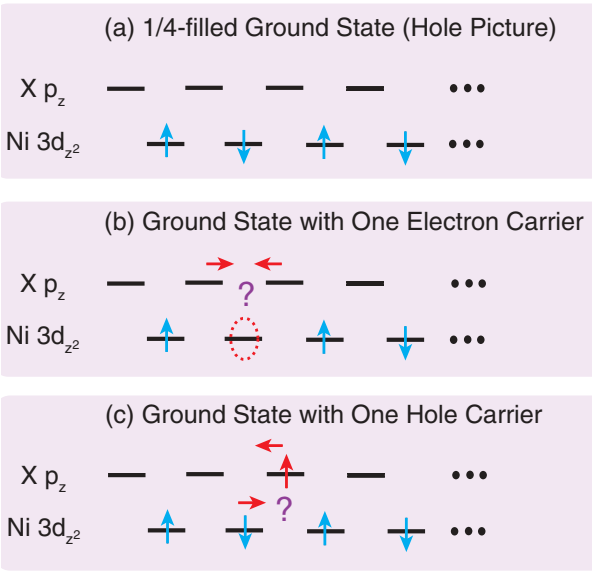

Figure 1
Hole arrangements in a NiX chain. The quarter-filled ground state is shown in (a), and those with one extra electron or one hole are shown *hypothetically* in (b) and (c). In (b) and (c), the horizontal arrows are also hypothetical movements of the ions.

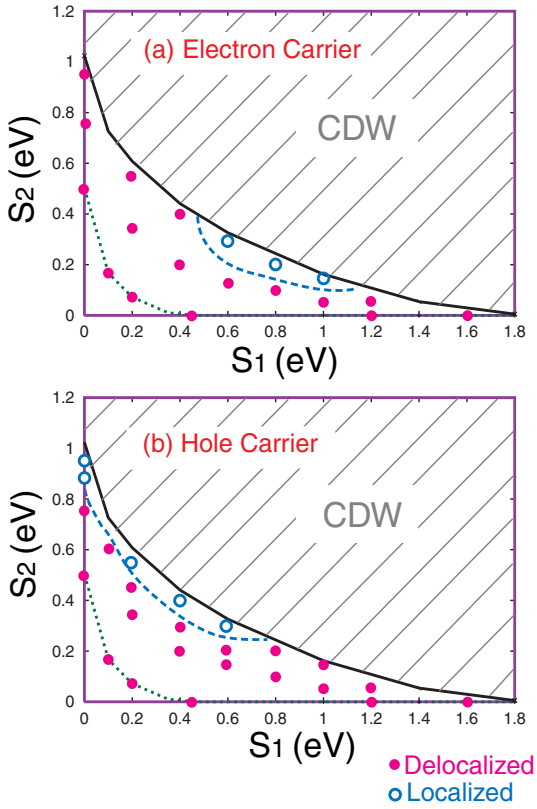

Figure 2
Phase diagram and summarized results for carrier localization and delocalization. S_1 (S_2) is α^2/K (β^2/K). The lower open region is the Mott-insulator phase that is our present concern. In this region, red closed (blue open) circles denote the points with a spatially delocalized (localized) carrier being stable. The upper shaded region corresponds to a charge-density-wave phase (CDW).

interaction of both the types represented by S_1 and S_2. This property is deeply related to the combination of the present type of e-l interactions and the strong electron correlation in the system. Namely, in the limit of strong U_d, eq. (1) is renormalized to a one-body problem of a holon (electron empty site) or a doublon (electron doubly occupied site). The present types of e-l interaction then give vanishing scattering rates to these particles. This contrasts well to the case of the original electrons in which the scattering rates remain finite.

We also discuss the relationship between our results and experiments. Firstly, an insulator-to-metal transition was observed for strong light intensity in a laser pump-probe experiment [3]. Such realization of the metallic state is considered to be basically explained in terms of our theory. For weak light intensity, a midgap absorption was observed. We assign this to a localization of electron carriers, presumably due to the distortion of the ligands or some disorder. Secondly, ARPES measurements for this material [4] showed no sign of polaronic effects. Since photoemission can be essentially related to a hole state, this fact is consistent with one of the present results, namely, the delocalization of holes.

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References

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