Spin and charge ordering in heterostructures of strongly correlated electron systems

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Abstract. In strongly correlated heterostructures, a non-uniform potential together with correlation effects can lead to novel electronic properties which are not realized in ordinary bulk systems. For example, it was reported experimentally and theoretically that the interface between the band insulator (BI) SrTiO$_3$ and the Mott insulator (MI) LaTiO$_3$ shows a metallic behavior. In this study, we consider a strongly correlated interface of BI and MI, like SrTiO$_3$/LaTiO$_3$, with particular emphasis on magnetic properties at the interface. To this end, we investigate the Hubbard model with long-range Coulomb interaction in the Hartree-Fock approximation. We find intriguing magnetic/charge phase transitions at the interface, which are closely related with the non-uniform potential. We elucidate that these transitions are caused by the strong coupling between charge and spin degrees of freedom near the interface. When the MI region has an antiferromagnetic (AF) order, a canted AF order can emerge around the interface, which shows a first-order metamagnetic transition under an external magnetic field. We also find that the strong spin-charge coupling stabilizes a charge order with a checkerboard pattern at the interface.

1. Introduction

Recent remarkable progress in crystal growth techniques has shed light on the frontier of the heterostructures composed of transition metal oxides. It is well known that electron correlation plays a crucial role in transition metal oxides. Therefore, in such heterostructures, we naturally expect the appearance of novel electronic phases due to the correlation effects under a non-uniform charge distribution. In pioneering experimental work, Ohtomo and co-workers investigated the correlated heterostructure made of different types of insulators such as SrTiO$_3$/LaTiO$_3$ (STO/LTO) and SrTiO$_3$/LaAlO$_3$ (STO/ LAO), and observed metallic conductivity [1, 2]. Furthermore, recent experiments reported a variety of properties at the correlated interfaces, which include superconductivity [3, 4], magnetic correlation [5], the effect of tunable Rashba spin-orbit interaction [6, 7], etc.

These experimental findings have stimulated intensive theoretical studies on strongly correlated layered systems [8–14]. For instance, Okamoto and Millis [9], and Rüegg et al. [11, 12] have investigated the correlation effects at the multilayered systems, and have clarified the characteristic metallic properties at the interface. Furthermore, Okamoto et al. [13], and Pentcheva and Pickett [14] have performed density functional calculations to investigate lattice relaxation effects.
In this paper, we systematically study the spin/charge properties characteristic for strongly correlated heterostructures. For this purpose, we investigate a generalized Hubbard model with the Hartree-Fock approximation. We first address a field-induced phase transition in external magnetic fields. It is found that the magnetization curve at the hetero-interface shows a first-order metamagnetic transition, the critical field of which is controlled by the strength of the on-site Coulomb interaction. This transition is accompanied by the modulation of the charge distribution near the interface. We also discuss other intriguing ordered states at zero magnetic field, such as a canted antiferromagnetic state and a charge ordered state, which are realized via the competition of the on-site Hubbard interaction and the long-range Coulomb interaction.

2. Model

We consider the strongly correlated heterostructure composed of Mott insulator (MI) sandwiched by band insulators (BI), like STO/LTO [1,4]. For simplicity, we neglect the orbital degeneracy. In order to reproduce the charge reconstruction in such multi-layered systems, we consider a generalized Hubbard model [9–12] given by

\[ H = H_{\text{band}} + H_U + H_{ee} + H_{ei}, \]  

where

\[ H_{\text{band}} + H_U = -t \sum_{<i,j>,\sigma} (c_i\sigma^\dagger c_j\sigma + \text{h.c.}) + U \sum_i \hat{n}_i\uparrow \hat{n}_i\downarrow, \]  

\[ H_{ee} + H_{ei} = \frac{1}{2} \sum_{i \neq j, \sigma, \sigma'} \frac{e^2}{|\vec{R}_i - \vec{R}_j|} \sum_{\epsilon l} \hat{n}_i\sigma \hat{n}_j\sigma', \]  

Here \( H_{\text{band}} + H_U \) \((H_{ee} + H_{ei})\) represents the ordinary single-band Hubbard model with nearest neighbor hopping \( t \) and on-site Coulomb repulsion \( U \) (long-range electron-electron and electron-ion interactions with dielectric constant \( \epsilon \) in the host lattice). In Eq. (3), \( \vec{R}_i = (l_i, m_i, n_i) \), with lattice constant \( a \), specifies a position of sites around which electrons move, and \( +e \) charged ions are placed at \( \vec{R}_{i,m} = a(l_i + 1/2, m_i + 1/2, n_i + 1/2) \). Note that the positions of the cations are confined within the MI region.

In the numerical analysis, open boundary conditions are adopted and the Hartree-Fock (Hartree) approximation is employed to treat the on-site (long range) electron-electron interactions. In the Hartree-Fock approximation, the on-site interaction term \( H_U \) in Eq. (2) is decoupled as follows,

\[ H_U \to U \sum_i \left\{ \langle \hat{n}_i\uparrow \rangle \hat{n}_i\downarrow + \langle \hat{n}_i\downarrow \rangle \hat{n}_i\uparrow - \langle \hat{n}_i\uparrow \rangle \langle \hat{n}_i\downarrow \rangle ight\} + \\
- \langle c_i\uparrow c_i\downarrow \rangle c_i\downarrow c_i\uparrow - \langle c_i\downarrow c_i\uparrow \rangle c_i\uparrow c_i\downarrow + \langle c_i\uparrow c_i\downarrow \rangle \langle c_i\downarrow c_i\uparrow \rangle \right\} \]  

where \( \langle \ldots \rangle \) denotes an expectation value within the mean-field treatment. In the following, we set \( a \) and \( t \) unity and restrict our investigation at zero temperature.

We note that the charge distribution in the heterostructure in this model is strongly affected by the strength of long-range Coulomb interaction \( E_c = e^2/\epsilon \) and the on-site repulsion \( U \) [11]. So, it is expected that the competition of these interactions leads to intriguing charge/spin phase transitions, which are inherent in the strongly correlated heterostructure. Therefore, we mainly focus on the effects of two interaction parameters \( U, E_c \), and investigate the ground-state properties of the interface.
3. Results and discussions

We present the computed results in this section. In the calculation, the strength of long-range Coulomb interaction is fixed at $E_c = 0.8$ for the heterostructure system with 25 layers, which includes 10 cation planes.

3.1. Metamagnetic transition

We start with the properties of the heterostructure under a magnetic field $\mathbf{H}$, which is applied perpendicularly to the direction of the antiferromagnetic (AF) moment realized in the MI region. To examine the effects of the external field, a Zeeman coupling term

$$H_Z = -\frac{1}{2} g \mu_B |\mathbf{H}| \sum_i m_{i,z},$$

is introduced in Eq. (1), where $g$ is the g-factor of electron, $\mu_B$ is the Bohr magneton and $m_{i,z} = \hat{n}_{i \uparrow} - \hat{n}_{i \downarrow}$ is the magnetic moment parallel to $\mathbf{H}$.

Figure 1 shows the behavior of the magnetization at the interface as a function of the magnetic field $H$ for a fixed value of on-site repulsion, $U = 8t$. Here, we define $H \equiv g \mu_B |\mathbf{H}|/2$ and $m_i \equiv \langle m_{i,z} \rangle$. With increasing $H$, it is clearly seen that the magnetization curve at the interface shows a discontinuous jump characteristic of first-order metamagnetic transition at $H \approx 0.825$, which is further confirmed by the existence of a hysteresis with decreasing $H$. On the other hand, we find that the magnetization at the central region of the MI state increases monotonically without exhibiting any jumps, thereby implying that such a metamagnetic behavior is intrinsic to the hetero-interface.

We show here that this metamagnetic transition is closely related to the charge localization effect due to the strong on-site interaction $U$. Figure 2 displays the $U$-dependence of the electron density at several layers, $l = 0$ (center layer), $l = 4$ (next to the interface) and $l = 5$ (interface) for $H = 0$. The gradients of the $l = 4$ and $l = 5$ curves change their signs at $U \sim 3t$, where the AF ordering appears. Similar behavior has previously been observed in the vicinity of a Mott transition point $U_c$ [11]. Ruegg et al. have suggested that the localization of the electronic states modifies the charge distribution for larger interactions beyond $U_c$, and electrons are confined to the MI region, i.e., the electron density at the interface decreases. Note that such a modification occurs only near the interface. All these behaviors are essentially the same as those observed here. Recall here that the external magnetic field has a tendency to suppress the AF ordering. This suppression is stronger at the interface than around center layers. This in turn increases the field-induced magnetic moment at the interface. In order to gain the energy due to the coupling with the magnetic field, the magnetically polarized interface has a tendency to increase the electron density, particularly in the large $U$-regime. We can, thus, say that the competition of $U$ and $H$ affects the AF ordering near the hetero-interface, giving rise to the metamagnetic

![Figure 1. Comparison of magnetization curves at the interface (open circle) and at the center of heterostructure (filled square). The up(down) arrow shows the behavior of the magnetization under the increase(decrease) of magnetic field. Here, the external magnetic field is applied perpendicular to the AF moment.](image-url)
transition which is accompanied by redistribution of electron charge near the interface. Actually, we have examined the same metamagnetic behavior in 2 ∼ 3 layers around the interface.

This view is indeed confirmed by our numerical results. Figure 3 shows that the metamagnetic behavior at the interface strongly depends on $U$. The metamagnetic transition is induced only for $U > U_M$ ($U_M \sim 8t$). In this regime, the critical magnetic field slightly decreases with increasing $U$. We also find that the metamagnetic transition is associated with the change in the electron density profile. We compute the electron occupation curve as a function of $H$, which exhibits a jump at the critical magnetic field corresponding to that of the metamagnetic transition. Moreover, a visible change in the electron density profile is found in the vicinity of the interface (not shown here). A detailed account of these results will be reported elsewhere.

3.2. Ground state phase diagram

Besides the above metamagnetism, we also elucidate here other intriguing phases at the correlated interface even without external magnetic fields. Figure 4 shows qualitatively the obtained $E_c − U$ phase diagram. We find a canted AF state and a charge ordered state with checkerboard pattern in the vicinity of the interface. In the following, we give a brief account of these states.

3.3. Canted AF state

Let us first mention a possible canted AF (CA) state. Considering the charge transfer, Yunoki et al. [15] have studied the competition or coexistence of different phases in the layers near hetero-
interfaces. Also, the previous dynamical mean field theory (DMFT) calculation performed by Okamoto and Millis [10] indicates a possible ordered state, which involves an AF ordering in central layers and a ferromagnetic (F) skin in the vicinity of the interface composed of BI and MI. Inspired by their studies, we here investigate the magnetic phase in the BI/MI/BI heterostructure. It is found that a CA state is induced by the interplay of AF and F ordering. Figure 5 describes the results obtained. Here, the on-site repulsion and the long-range interaction are fixed at $U = 11t$ and $E_c = 0.8t$, respectively. As suggested in earlier work [10], we observe the coexistence of the AF and F states in the heterostructure (Fig. 5). More interestingly, a close examination of this state shows that the F moment lies perpendicular to the AF moment, and, thus, a CA state is realized near the interface. More detailed studies of this magnetic properties are in progress.

3.4. Charge ordered state

In order to understand the effect of charge localization, which may be caused by the competition of $E_c$ and $U$ [11], the $E_c - U$ phase diagram is investigated. A charge-ordered (CO) state with checkerboard pattern is found in the region above $E_c \sim 1.0$. Pentcheva and Pickett previously pointed out the existence of the CO phase with ab initio calculations [14], and showed that the $d_{xy}$ orbital induces the CO at the interface. Thus, our present result is conceptually consistent with their finding. Here, we give a close investigation on the origin of the CO state to address the role of $E_c$ and $U$.

In Fig. 6, we plot the order parameter of the CO state as a function of $E_c$. The effect of the interlayer hopping $t_z$ is investigated in the isotropic case (open circle) and the anisotropic case (filled square).
density at the interface is $0.4 \sim 0.5$, close to quarter filling [14]. However, another mechanism may be necessary to explain the CO state for $E_c < 1.5$. For the realization of the CO state in this region, the magnetic interaction between the interface and the MI region plays an important role. In such a parameter region, the inhomogeneity of charge distribution in the interfacial plane can be induced by the interlayer spin coupling; some sites (lower occupancy) prefer the F ordering with their neighboring layers, while the other sites (higher occupancy) prefer the AF one. This enhances a tendency to form the in-plane CO state even for relatively weak $E_c$. We can thus say that the formation of the CO state is an intrinsic property in the hetero-interface.

4. Summary

We presented the mean field study of the spin/charge ordering in the strongly correlated heterostructure. We found the first-order metamagnetic transition in the vicinity of the interface. The resulting magnetization curve shows a characteristic jump-structure accompanied by a hysteresis. It has been shown that such a transition is caused by the competition of correlation ($U$) and the uniform spin polarization by $H$. This mechanism is indeed fully consistent with the $U$ dependence of the metamagnetic transition and the jump in the electron density as a function of $H$. We have also addressed other interesting interfacial states which are realized in the absence of magnetic fields. We have confirmed the interfacial state suggested in previous work [10] and found the appearance of canted antiferromagnetism. Furthermore, we have found the existence of a charge ordered state even for weak Coulomb interactions, which is stabilized by the spin and charge coupling in the vicinity of the hetero-interface.

Since the present investigation has been done within the mean-field treatment, it is important to confirm the results by taking into account strong electron correlations with many-body techniques such as dynamical mean field theory. This issue is now in progress, and the detailed results will be reported elsewhere.

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