The existence and origin of “striped” structures continues attracting considerable attention in the research field of transition-metal oxides. In a system with dominant electron-electron repulsion, the Wigner-crystal state should be stabilized, but in real materials more complicated non-uniform charge structures have been found. In Nd-based lightly-doped cuprates, neutron scattering experiments revealed incommensurate spin structures, where antiferromagnetic (AFM) spin stripes are periodically separated by domain walls of holes. In La$_2$−$x$Sr$_x$CuO$_4$, dynamical stripes are believed to exist along vertical or horizontal directions (Cu-O bond direction). In nickelates, the charge-ordered stripes are along the diagonal direction. In manganites, evidence for striped charge-ordering also along the diagonal direction has been reported in the AFM phase for $x>1/2$, while short-range diagonal stripe correlations have been found in the ferromagnetic (FM) phase at $x<1/2$.

In general, stripes can be classified into metallic or insulating. In La$_{2-x}$Sr$_x$CuO$_4$, the dynamical stripes exhibit metallic properties, but they are easily pinned by lattice effects and impurities. In La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$, stripes along the bond-direction are pinned by lattice distortions, but they are still metallic. Intuitively, vertical or horizontal stripes could be associated with the formation of “rivers of holes”, to prevent individual charges from fighting against the AFM background. Such stripes should be metallic, even if they are pinned, since they are induced by the optimization of hole motion between nearest-neighbor Cu-sites via oxygens.

However, in the diagonal stripes observed in manganites and nickelates, charges are basically localized, indicating that such insulating stripes are not determined just by the optimization of the hole motion. In the FM state of manganites, the hole movement is already optimal and, naively, charges should not form stripes. Obviously, an additional effective local potential must be acting to confine electrons into stripes. If such a potential originates in lattice distortions, it is expected to occur along the bond direction to avoid energy loss due to the conflict between neighboring lattice distortions sharing the same oxygens. Then, static stripes stabilized by lattice distortions tend to occur along the diagonal direction, as shown in the stripes of the FM-phase of manganites, stabilized by Jahn-Teller (JT) distortions.

In simple terms, vertical or horizontal stripes in cuprates can be understood by the competition between Coulomb interaction and hole motion, while diagonal stripes are better explained as a consequence of a robust electron-lattice coupling. However, a difficulty has been found for theoretical studies of stripe formation in doped nickelates, since both Coulomb interaction and electron-lattice coupling appear to be important. Since the Ni$^{2+}$ ion has two electrons in the $e_g$ orbitals, on-site Coulomb interactions certainly play a crucial role to form spins $S=1$. When holes are doped, one electron is removed and another remains in the $e_g$ orbitals, indicating that the hole-doped site should become JT active. Then, in hole-doped nickelates both Coulombic and phononic interactions could be of relevance, a fact not considered in previous theoretical investigations.

In this Letter, charge ordering in doped nickelates is investigated based on the orbital-degenerate Hubbard model coupled to lattice distortions, using numerical techniques. After confirming the Néel state composed of $S=1$ spins at $x=0$, both cases $x=1/2$ and $1/3$ will be analyzed. At $x=1/2$, novel AFM phases called CE- and E-type have been unveiled, which are consistent with experimental results. Further including the JT-type cooperative distortion, $(3x^2−r^2/3y^2−r^2)$-type orbital ordering is predicted. For $x=1/3$, an incommensurate spin structure is induced by the Coulombic model, including the level splitting between $e_g$ orbitals, but the charge stripe does not appear. To reproduce simultaneously spin and charge stripes, it is important to include the strong coupling of $e_g$ electrons to lattice distortions, originating in the in-plane oxygen motions.

The model for nickelates includes three important ingredients: The kinetic motion of $e_g$ electrons, Coulomb interactions among $e_g$ electrons, and electron-lattice cou-
plittings between $e_g$ electrons and distortions of the NiO$_6$ octahedra. Note that the electron-lattice term is divided into couplings for the apical and in-plane oxygen motions. In layered nickelates, all NiO$_6$ octahedra are significantly elongated along the c-axis, splitting the $e_g$ orbitals. This splitting from apical oxygen should be included explicitly from the start and, then, the in-plane motion should be studied. The Hamiltonian $H$ is given by

$$H = -\sum_{i\gamma\gamma'} t_{\gamma\gamma'}^{\pi} d_{i\gamma\sigma}^\dagger d_{i+\gamma'y'\sigma} + \Delta \sum_i (n_{ia} - n_{ib})/2$$
$$+ U \sum_i n_{i\pi\uparrow} n_{i\pi\downarrow} + J \sum_{i,\sigma',\sigma} d_{i\sigma\gamma}^\dagger d_{i\sigma'\gamma} d_{i\sigma\gamma'}^\dagger d_{i\sigma'\gamma'}$$
$$+ U' \sum_i n_{ia} n_{ib} + J' \sum_{i,\gamma\gamma'} d_{i\gamma\uparrow}^\dagger d_{i+1\gamma\uparrow} d_{i\gamma'\downarrow} d_{i+1\gamma'\downarrow}, \tag{1}$$

where $d_{i\sigma}$ ($d_{ib\sigma}$) is the annihilation operator for an $e_g$-electron with spin $\sigma$ in the $d_{x^2-y^2}$ ($d_{3z^2-r^2}$) orbital at site $i$, $n_{i\gamma\sigma} = d_{i\gamma\sigma}^\dagger d_{i\gamma\sigma}$, $n_{ia} = \sum_{\gamma} n_{i\gamma\sigma}$, $a$ is the vector connecting nearest-neighbor sites, and $t_{\gamma\gamma'}^{\pi}$ is the nearest-neighbor hopping amplitude between $\gamma$- and $\gamma'$-orbitals along the $a$-direction, given by $t_{aa}^{\pi} = -\sqrt{3}t_{ba}^{\pi} = -\sqrt{3}t_{bb}^{\pi} = -3t/4$ for the $x$-direction and $t_{ba}^{\pi} = \sqrt{3}t_{ab}^{\pi} = 3t/4$ for the $y$-direction. Hereafter, $t$ is the energy unit. In the second term, $\Delta > 0$ is the level splitting between $a$- and $b$-orbitals. In the Coulomb interaction terms, $U$ ($U'$) is the intra- orbital (inter- orbital) Coulomb interaction, $J$ is the inter-orbital exchange interaction, and $J'$ is the pair-hopping amplitude between different orbitals. Due to the relations $J = J'$ and $U = U' + J + J'$, the independent parameters are $U'$ and $J$, with $U' > J$. The calculations are all carried out using standard exact-diagonalization techniques. It is crucial to use this kind of unbiased methods for this first study that includes both Coulomb and lattice effects, even if the technique restricts us to small $N$-site clusters.

First, consider the undoped case. The calculation is done for an 8-site tilted cluster, equivalent in complexity to a 16-site lattice for the single-band Hubbard model. Since at all sites the two orbitals are occupied due to the Hund’s rule coupling, the JT distortions are not active and it is possible to grasp the essential ground-state properties using $H$. In Fig. 1(a), the Fourier transform of spin correlations is shown vs. $\Delta$, where $S(q) = (1/N) \sum_{ij} e^{iq(i-j)} \langle S_i^q S_j^q \rangle$, with $S_i^q = \sum_{\gamma} \langle d_{i\gamma\uparrow} d_{i\gamma\uparrow} - d_{i\gamma\downarrow} d_{i\gamma\downarrow} \rangle/2$. As expected, a robust $\pi \neq 2\pi$ peak can be observed for $\Delta \lesssim 3$, suggesting that the AFM phase is stabilized by super-exchange interactions. The rapid decrease of $S(\pi, \pi)$ for $\Delta > 3$ is understood by comparing the energies for local triplet and singlet states, as shown in Fig. 1(b). The ground-state properties change at $U' - J = U - \Delta$, leading to $\Delta = 3J$ for the transition. The spin structure at $x = 0$ is schematically shown in Fig. 1(c).

Let us turn our attention to the case $x = 1/2$. The 8-site tilted lattice is again used for the analysis, and the phase diagram Fig. 2(a) is obtained for $\Delta = 0.5$. Increasing $J$, an interesting transformation from AFM to FM phases is found. This is natural, since at large $J$ the system has a formal similarity with manganite models, where kinetic-energy gains lead to ferromagnetism, while at small $J$ the magnetic energy dominates. However, between the G-type AFM for $J = 0$ and FM phase for $J = U'$, unexpected states appear which are mixtures of FM and AFM phases, due to the competition between kinetic and magnetic energies. Typical spin correlations $S(q)$ are shown in Fig. 2(b). Note that peaks at $q = (\pi, 0)$ and $(\pi/2, \pi/2)$ indicate “C” and “E” type spin-structures, respectively (the notation is borrowed from the Mn-oxide context). Double peaks at $q = (\pi, 0)$ and $(\pi/2, \pi/2)$ denote the CE-type structure, frequently observed in half-doped manganites. In half-doped nickelates, the CE-phase is expressed as a mixture of type (I) and (II) in Fig. 2(c), depending on the positions of the $S = 1$ and $S = 1/2$ sites, although the “zigzag” FM chain structure is common for both types. The E-type phase is also depicted in Fig. 2(c). Note that the charge correlation always exhibits a peak at $q = (\pi, 0)$ (not shown here), indicating the checkerboard-type charge ordering.

In experimental results, a peak at $(\pi/2, \pi/2)$ in $S(q)$ has been reported, suggesting an AFM pair of $S = 1$ spins across the singly-occupied sites with holes. Moreover, the checkerboard-type charge ordering has been experimentally observed. Thus, the spin-charge patterns of CE(II)- and E-type are consistent with the experimental results. Our phase diagram has a robust region with a peak at $(\pi/2, \pi/2)$, both for CE- and E-type phases, although the CE-phase exhibits an extra peak at $(\pi, 0)$. Whether the E- or CE-phases are present in nickelates can be studied experimentally in the future by searching for this $(\pi, 0)$ peak. Note that if diffuse scattering experiments detect the AF correlation along the hole stripe, as has been found at $x = 1/3$, the CE(II)-type may be the only possibility. Summarizing, the spin-charge structure in $x = 1/2$ experiments can be understood within the Hamiltonian $H$ by assuming a relatively large $J$. 

FIG. 1: (a) Spin correlation $S(q)$ vs. $\Delta$ for $x = 0$. (b) Two kinds of local $e_g$-electron arrangements for $x = 0$. (c) AFM spin pattern theoretically determined for $\Delta \lesssim 3$. 
Consider now the effect of in-plane oxygen motion (apical oxygen motions have already been included as an \( \epsilon_{g} \)-level splitting). Assuming that oxygens move along the Ni-O bond direction, the extra electron-phonon term is written as

\[
H_{\text{eph}} = g \sum_{i} \left[ -Q_{1i}(n_{i\sigma} + n_{i\bar{\sigma}}) + Q_{2i} \tau_{z,i} + Q_{3i} \tau_{x,i} \right] + (k/2) \sum_{i} (\beta Q_{1i}^2 + Q_{2i}^2 + Q_{3i}^2),
\]

where \( g \) is the electron-lattice coupling constant, \( Q_{1i} \) is the breathing-mode distortion, \( Q_{2i} \) and \( Q_{3i} \) are, respectively, the \( (x^2-y^2) \) - and \( (3z^2-r^2) \)-type JT distortions, \( \tau_{z,i} = \sum_{\sigma}(d_{i\sigma}^a d_{i\bar{\sigma}} + d_{i\bar{\sigma}}^a d_{i\sigma}) \), and \( \tau_{x,i} = \sum_{\sigma}(d_{i\sigma}^a d_{i\sigma} - d_{i\bar{\sigma}}^a d_{i\bar{\sigma}}) \). The second term is the quadratic potential for adiabatic distortions, where \( k \) is the spring constant for the JT-mode and \( \beta \) is the spring-constant ratio for breathing- and JT-modes. From our experience in manganites, this ratio is here fixed to \( \beta=2 \).

Since all oxygens are shared by adjacent NiO\(_6\) octahedra, the distortions are not independent. To consider such cooperative effect, in principle, the O-ion displacements should be optimized. However, in practice it is not feasible to perform both the Lanczos diagonalization and the optimization of all oxygen positions for 6- and 8-site clusters. In the actual calculations, \( Q_{1i} \), \( Q_{2i} \), and \( Q_{3i} \) are expressed by a single parameter \( d \), for the shift of the O-ion coordinate. Note that the unit of \( d \) is \( g/k \), typically \( 0.1-0.3 \)Å. Then, the total energy is evaluated as a function of \( d \) to find the minimum energy state. Repeating these calculations for several distortion patterns, it is possible to deduce the optimal state.

After several trials, the optimal distortion at \( x=1/2 \) is shown in Fig. 2(d). The diagonalization has been performed at several values of \( d \) on the 8-site distorted lattice and the minimum in the total energy is found at \( d=0.3 \) (Fig. 2(e)). Here, the dimensionless coupling constant \( \lambda \) is defined as \( \lambda=g/\sqrt{k} \). As mentioned above, even without \( H_{\text{eph}} \), the checkerboard-type charge ordering has been obtained, but the peak at \( q=(\pi, \pi) \) significantly grows due to the effect of lattice distortions. Note that the distortion pattern in Fig. 2(d) is essentially the same as that for half-doped manganites. This is quite natural, since JT and active ions exist bipartitely also for half-doped nickelates. Then, due to this JT-type distortion orbital ordering for half-doped nickelates is predicted, as schematically shown in Fig. 2(d). The shapes of orbitals are determined from the orbital densities, \( \langle \tau_{z,i} \rangle \) and \( \langle \tau_{x,i} \rangle \) (Fig. 2(f)). The well-known alternate pattern of \( 3x^2-r^2 \) and \( 3y^2-r^2 \) orbitals in half-doped manganites is denoted by dashed lines. Increasing \( d \), the shape of orbitals deviates from \( 3x^2-r^2 \) and \( 3y^2-r^2 \), but it is still characterized by the orbitals elongating along the \( x \)- and \( y \)-directions (see insets of Fig. 2(f)). It would be very interesting to search for orbital ordering in half-doped nickelates, using the resonant X-ray scattering technique.

Now let us move to the case \( x=1/3 \). If the actual expected stripe structure at \( x=1/3 \) is faithfully considered, it is necessary to analyze, at least, a \( 6 \times 6 \) cluster. However, such a large-size cluster with orbital degeneracy cannot be treated exactly due to the exponential growth of the Hilbert space with cluster size. Then, a covering of the two-dimensional (2D) lattice using zigzag 6-sites clusters is considered (Fig. 3(a)) by assuming a periodic structure along the diagonal direction. The phase diagram obtained by analyzing the zigzag 6-site cluster for \( H \) is in Fig. 3(b). Typical spin and charge correlations are in Figs. 3(c) and (d), where \( C(q)=(1/N)\sum_{i,j} e^{iq \cdot (i-j)} \langle n_i - \langle n \rangle \rangle \langle n_j - \langle n \rangle \rangle \), with \( n_i = \sum_{\sigma} \delta_{i\sigma} \). Since the momentum \( q \) is defined along the zigzag direction, the phase labelled by \( q=2\pi/3 \) in Fig. 3(b) denotes an incommensurate AFM phase with the proper spin stripe structure. The phase labelled by \( q=\pi/3 \) indicates...
a spin spiral state, which will eventually turn to the FM phase in the thermodynamic limit. Thus, the spin stripe phase appears between the commensurate AFM and FM-like phases, similar to the case of $x=1/2$. However, as seen in Fig. 3(d), $C(q)$ in the spin stripe phase does not show the striped charge structure ($q=2\pi/3$). Rather, bipartite charge ordering characterized by a peak at $q=\pi$ still remains. Namely, the Hamiltonian $H$ can explain the spin stripe, but does not reproduce the striped charge ordering at $x=1/3$, indicating the importance of $H_{eph}$.

Consider now the effect of $H_{eph}$ for $x=1/3$. After evaluating total ground-state energies for several kinds of distortions, the pattern in Fig. 3(e) has been found to provide the optimal state at $x=1/3$. This type of distortion induces a spatial modulation of the level splitting as $-\delta_1/2=\delta_2=\delta_3=-\delta_4/2=\delta_5=\delta_6$, where $\delta_1$ is the level splitting caused by the in-plane oxygen motions, and the site numbers are in Fig. 3(e). The minimum energy is found at $d=0.1$ (Fig. 3(f)). The modulation of level splitting stabilizes the striped charge ordering characterized by a $q=2\pi/3$ peak in $C(q)$ (Fig. 3(g)).

Note that $(3x^2-r^2/3y^2-r^2)$-type orbital ordering does not occur in Fig. 3(e). Phenomenologically, such orbital ordering tends to appear in a hole pair separated by one site, the unit of the “bi-stripe” of manganites. However, such a bi-stripe-type ordering contradicts the $x=1/3$ striped charge-ordering, and the bi-stripe-type solution was found to be unstable in these calculations. One may consider other distortion patterns which satisfy both $(3x^2-r^2/3y^2-r^2)$-type orbital and striped charge-ordering, but in such distortions no energy minimum was obtained for $d>0$. After several trials, Fig. 3(e) has provided the most optimal state.

Summarizing, possible spin, charge, and orbital structures of layered nickelates have been discussed based on the $e_g$-orbital degenerate Hubbard model coupled with lattice distortions. To understand the nickelate stripes, both Hund’s rule interaction and electron-lattice coupling appear essentially important. At $x=1/2$, $(3x^2-r^2/3y^2-r^2)$-type orbital ordering similar to that in half-doped manganites is predicted. Even FM phases could be stabilized by chemically altering the carrier’s bandwidth. For $x=1/3$, a spatial modulation in level splitting plays an important role for stripe formation.

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