A disparate variety of interacting models based on density functional theory (DFT) studies have been proposed to illuminate the electronic and magnetic behavior underlying the discovery of long-sought superconductivity in a layered nickelate, specifically hole-doped NdNiO$_2$ (NNO). With the same infinite-layer structure as CaCuO$_2$ (CCO), which superconducts up to 110 K when doped, NNO displays several differences with CCO. Undoped NNO is conducting; CCO is insulating; CCO orders antiferromagnetically, whereas no signature of order is seen in NNO (and its iso-electronic but non-superconducting sisterLaNiO$_2$ (LNO)) and there is no sign of heavy feronm screening of Ni moments. CCO and NNO have a common $d^9$ (formal) configuration (one hole) on the transition metal ion, however NNO persists as conducting to low temperature. Central differences include positioning of metal 3d levels relative to O 2p levels that is substantially different in the two compounds, and the presence of Nd 5d character at the Fermi level.

The electronic structures of the $d^9$ infinite layer cuprates and nickelates were compared some time ago, with some similarities but substantial differences being noted and quantified more recently by Wannier function analysis, and some impact of the Nd moments has been noted. The similarities but substantial difference continue to dominate the discussion and inform the models cited above. No items however seem as fundamental as the difference in ground states of the undoped materials: antiferromagnetic (AFM) insulator for CCO, and disordered moment conductor for NNO, the latter being a conducting quantum paramagnetic (MQPM) phase in the Sachdev-Read classification. Based on an interacting two-band model, Werner and Hoshino discussed NNO in terms of the spin-freezing theory of unconventional superconductivity, and the potentially simpler case of superconductivity at high pressure in elemental Eu with disordered f$^7$ moments might share in this behavior.

Here we focus on this absence of (AFM) ordering of Ni moments in NNO, moments whose ordering has been the overriding hallmark of the undoped phase in cuprates. In closely related layered nickelates $\text{LaNiO}_2$ and $\text{La}_2\text{Ni}_2\text{O}_4$ (viz. $\text{La}_2\text{Ni}_2\text{O}_4$, $\text{La}_2\text{Ni}_3\text{O}_8$, $\text{La}_2\text{Ni}_3\text{O}_{10}$) even with non-integral formal Ni valences magnetic ordering and sometimes charge ordering occur; none has a ground state without symmetry breaking, unlike NNO. In spite of being energetically favored in the absence of fluctuations (i.e. in DFT calculations), the ordered state (with $S=1$ quantum fluctuations) is not observed, hence it is a metastable but physically inaccessible phase. The AFM state of NNO is however accessible computationally, allowing study of its electronic structure and microscopic processes that oppose further symmetry breaking, thereby giving insight into the inaccessibility of the AFM phase.

We adopt the DFT plus anisotropic Coulomb $U$ (DFT+$U$) method as implemented in Wien2k as described in our previous work, retaining the all-electron character of Nd which provides Nd $d-f$ exchange coupling but is not the subject here. Details of the frozen phonon studies are provided in the Supplemental Material (SM). We use Hund’s exchange coupling $J_{H}^{\text{Hund}} = 0.7$ eV throughout, and quote values of $U_N$ (simply called $U$) that we explore.

**Energies and moments.** The AFM state we study has both antialigned Nd and Ni spin layers, with no net near neighbor Nd-Ni coupling. This AFM ordering is energetically favored over aligned Ni spins by 95 (62) meV/f.u. at $U = 0(4)$ eV, both being strongly favored over non-magnetic Ni. No theoretical value of the energy for disordered moments is available, but would be of the order of $J_{H}^{\text{Hund}}$. Although DFT often overestimates magnetic moments and thereby energies in weak magnets, it is
commonly accurate for larger (ionic) moments. These Ni moments are \( \sim 1 \mu_B \) or more, characteristic of hybridized S=1/2 moments but only if hybridization with oxygen is strong or a second orbital is involved. Adding the effect of \( U \) renews the energetics and increases the moments, but not the relative stabilities of AMF and FM order. The observed avoidance by NNO of a relatively large ordering parameter is \( t_{zz} \approx 1 \) eV, large for \( k_z \) hopping across a 10Å distance. For the minority (higher energy) states, the Ni \( d_{zz} \) orbital is mixed with Nd \( d_{xy} \) and somewhat with Nd \( d_{x^2-y^2} \) on the \( k_z = \pi/c \) plane, resulting in a hybridization gap of 0.4 eV at the \( Z \)-point. The mixtures elsewhere, viz. at \( \Gamma \), are negligible.

Figure 2 shows (for the realistic metallic value \( U=4 \) eV) Ni, Nd, and O orbital-projected densities of states (PDOSs) in the \( d \) band region surrounding \( E_F \). The unfilled minority Ni \( d_{zz} \) orbital (the hole) lies at +3 eV. One remarkable feature is that the minority Ni \( d_{zz} \) band spans nearly 5 eV, crossing \( E_F \) and ensuring a conducting state. All other Ni \( d \) orbitals of both spins are narrow. The other notable feature is that this same orbital gives rise to a flat band across the entire \( k_z = \pi/c \) zone face that produces a 1D van Hove singularity (vHs) \( \text{pinned at } E_F \), with pure \( d_{zz} \) character; recall, these two exotic features are robust, that is, insensitive to \( U \). The valence bands have nearly pure Ni \( d_{zz} \) character within 1 eV of \( E_F \). Nd \( 5d \) character extends down to and slightly below \( E_F \) as noted several times previously, but does not participate in the flat band. Around \( Z \) (see along the \( Z-R \) and \( Z-A \) lines in Fig. 2), the flat band crosses a dispersive band with mixed Nd \( 5d_{zz} \), Nd \( 3d_{x^2-y^2} \) character at \( E_F \).

As a result of coupling through Nd states, the flat band acquires a small width \( \sim 40 \) meV corresponding to an effective \( \text{in-plane hopping } t_{zz}^{zz} = 20 \) meV for this plane. Based on charge decompositions and PDOSs, and the Ni \( d_{zz} \) character above \( E_F \), the evidence suggests a formal valence closer to Ni\(^{1.4+}\) rather than Ni\(^{1+}\) as would be appropriate for an insulator. (Since O is clearly 2−, we expect this is a breakdown of formal valence counting as

FIG. 1: Fatband depiction of the AFM band structure in GGA+U: (Left) Ni majority; (right) Ni minority. Panels are for \( U=0, 1, 2 \) eV as noted, with \( E_F = 0 \). The flat \( d_{zz} \) band appears along the \( Z-R-A-Z \) lines (\( k_z = \pi/c \) zone face) already for \( U=1 \) eV. Ni \( d_{zz} \), \( d_{x^2-y^2} \), and \( d_{xy} \) characters (described in the primitive cell) are highlighted by brown, pink, and green colors, respectively. Relative to the primitive cell, symmetry points are rotated by 45° in the \( \sqrt{2} \times \sqrt{2} \) AFM supercell, i.e., \( M(R) \leftrightarrow X(A) \).

FIG. 2: AFM orbital-projected densities of states (PDOSs)/atom of Ni 3d, two Nd 5d \( d_{zz} \) and \( d_{xy} \) orbitals, and O 2p orbitals, for the undistorted lattice at \( U = 4 \) eV, in the 3d band region (note the difference in scales). Majority (minority) are plotted upward (downward). The oxygen PDOS lies below 3 eV. Having \( E_F \) pinned precisely at the sharp, 1D-like vHs (upper panel, minority spin, shown enlarged in the Inset) produces the instabilities discussed in the text.
The virtual crystal approximation was used to replace the atomic number of Nd. Often occurs in metals.) In contrast to cuprates where the central role implicates the Cu \(d_{x^2-y^2}\) orbital, our results support previous indications\[15\] that the Ni \(d_{x^2}\) orbital becomes a central player in NNO (and LNO), with the \(d_{x^2-y^2}\) hole being more of a given and less of a dynamical component.

Fermi surfaces versus doping. The Fermi surfaces (FSs) for \(U = 4\) eV versus small (virtual crystal) doping levels are shown in Fig. 3. A \(\Gamma\)-centered electronic sphere, a mixture of Ni \(3d_z^2\) and Nd \(5d_z^2\) character,\[15, 19\] arises from the high velocity band. Quasi-1D pieces emerge on the \(k_z = \pi/c\) plane. For hole-doping, the thin hole wafer is pinched off around the \(Z\)-point (Fig. 3(a)). At stoichiometry, Fig. 3(b), the hole FS shrinks and flat electron buttons appear around the \(Z\) point, leaving rounded-diamond hole buttons at the zone vertices. Small electron doping, Fig. 3(c), leads to electron buttons around \(Z\); the electron sphere at \(\Gamma\) is simply changed in volume during low doping. The thin button FSs have a characteristic thickness \(\Delta k_z \leq 0.06\pi/c\) or less, providing nesting that would encourage charge- or spin-density waves (or related Kohn anomalies) of wavelength greater than \(2\pi/\Delta k_z \sim 8c\), supported by nesting extending over a range of parallel \(q\) components. Such long wavelength fluctuations are difficult numerically to explore.

Magnetic Instability. To probe the Stoner instability in this already magnetically-ordered AFM state, we use lattice displacements [Fig. 4(a)] to break symmetry. The changes in moments of the Ni1 and Ni2 sites are displayed in Fig. 4(b). Unexpectedly, tiny mode amplitudes (displacements down to 0.001\(\text{Å}\) extrapolating to zero) produce a ‘spin disproportionation’ of the order of ±0.05 \(\mu_B\). To clarify: the AFM state is unstable to a first order transition to a ferrimagnetic state, which reflects the partially itinerant character of the Ni moments. The mechanism is the vHs/Stoner instability, with feedback within the self-consistency loop (necessitated by the vHs) providing the first order stabilization; actual lattice distortion is unnecessary. The magnetic symmetry breaking is accompanied by a Ni1-to-Ni2 charge transfer up to \(\sim \pm 0.05e\) (from atomic sphere charges).

Lattice Instability. The effect of the breathing mode on the band structure is displayed in Fig. 5. The (red and green) dashed lines indicate the bands split by the full-breathing mode, with O displaced by 0.03\(\text{Å}\). The half-breathing mode (not shown) shows similar band splitting. The peak-splitting is the source of the Peierls (Stoner) charge (spin) instability.
adiabatic electron-lattice coupling, where heavy charge carriers are coupled to phonons of similar energy.

However, there is oxygen zero point motion to account for. For frequencies of 60-80 meV, the oxygen zero point amplitude $\langle u_o \rangle \approx 0.06 \text{Å}$ is four times larger than the region of non-adiabatic behavior (Fig. 5), so this quantum fluctuation will frustrate the static lattice instability, and quench the spin and charge orders as well. How the massive carriers would deal with all of these frustrated instabilities promises to be a challenging theoretical problem, but the outcome is that the underlying AFM order we have modeled gives way to a disordered moment metal.

Discussion: avoidance of the AFM phase. We have studied the experimentally inaccessible AFM ordered state of NdNiO$_2$ with correlated DFT methods. This is the state whose calculated energy indicates it should be the ground state versus the observed disordered moment state. We found that a Ni $d_{xz}$ flat band arises on the entire $k_z = \pi/c$ zone face, giving a 1D-like vHs that supports (neglecting fluctuations) spin, charge, and lattice (breathing and half-breathing mode) instabilities of the ideal infinite-layer lattice. Due to the narrowness of the vHs, these coupled order parameters are sensitive to quantum zero point oxygen motion and non-adiabatic electron-lattice coupling in the vHs peak, finally frustrating these types of lowering of symmetry.

Yet the AFM ordered state, calculated to be most stable for the static lattice, remains inaccessible by experiment, $i.e.$ higher in free energy than the spin-disordered, spin liquid state. We propose that as temperature is lowered NdNiO$_2$ approaches the AFM ordered phase but encounters its incipient instabilities with strong spin and charge fluctuations that inhibit spin order. The result is to remain spin-disordered but with strong long range correlations, $i.e.$ incipient AFM order, that reduce the free energy. This correlated spin liquid phase, the Sachdev-Read MQPM phase that has been proposed in another disordered moment superconductor, promises to provide the platform for the superconductivity that appears upon hole doping.

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FIG. 6: Energy distortions for the frozen-in oxygen full- and half-breathing modes at $U = 4$ eV. As shown in the Inset, these follow a quadratic plus lowest-order anharmonic term, indicated by the green (blue) dashed lines, and are insensitive to strength of $U$ in the range of 3 - 5 eV.

The anomalous response is confined to small distortions and energies (and temperature). Figure 6 shows a deformation potential: peak splitting per unit $\omega$ (4E/$u^2$) in the range of 3 - 5 eV.

The sharp and narrow vHs however negates normal charge response and provides a platform that is extremely sensitive to any symmetry breaking that splits the vHs peak. The anomalous response is confined to small distortions and energies (and temperature). Figure 6 shows that as the amplitude $u$ is decreased below 0.015 Å, the lattice stiffness diverges to lower values and extrapolates to negative values below $u = 0.004$ Å. This is the classic Peierls instability, and a full treatment (unnecessary here) would require self-consistent theory of non-adiabatic electron-lattice coupling, where heavy charge

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