Finite-temperature properties of the triangular lattice $t$-$J$ model and applications to Na$_x$CoO$_2$

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We present a finite temperature ($T$) study of the $t$-$J$ model on the two-dimensional triangular lattice for the negative hopping $t$, as relevant for the electron-doped Na$_x$CoO$_2$ (NCO). We study several thermodynamic and transport properties in this study: the $T$-dependent chemical potential, specific heat, magnetic susceptibility, and the dynamic Hall coefficient across the entire doping range. We show systematically how this simplest model for strongly correlated electrons describes a crossover as function of doping ($x$) from a Pauli-like weakly spin-correlated metal close to the band limit (density $n=2$) to the Curie-Weiss metallic phase ($1.5<n<1.75$) with pronounced antiferromagnetic (AFM) correlations at low temperatures and Curie-Weiss-type behavior in the high-temperature regime. Upon further reduction of the doping, a different energy scale, dominated by spin-interactions ($J$) emerges. It is apparent both in specific heat and susceptibility, and we identify an effective interaction $J_{\text{eff}}(x)$, valid across the entire doping range. This is in contrast to the formula by Anderson et al. [J. Phys.: Condens. Matter 16, R755 (2004)] for the square lattice. NCO has $t<0$, hence the opposite sign of the Nagaoka-ferromagnetic situation, this expression includes the subtle effect of weak kinetic AFM [Haerter and Shastry, Phys. Rev. Lett. 95, 087202 (2005)], as encountered in the infinitely correlated situation ($U=\infty$) for electronic frustration. By explicit computation of the Kubo formulas, we address the question of practical relevance of the high-frequency expression for the Hall coefficient $R_H$ [Shastry et al., Phys. Rev. Lett. 70, 2004 (1993)]. We hope to clarify some open questions concerning the applicability of the $t$-$J$ model to real experimental situations through this study.

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I. INTRODUCTION

Since the discovery of superconductivity for $x \approx 0.3$ in H$_2$O-intercalated Na$_x$CoO$_2$, or NCO crystals, this compound has drawn significant attention. NCO also displays a variety of remarkable thermoelectric effects, such as an unusually large and magnetic-field dependent thermopower for a variety of remarkable thermoelectric effects, such as an unpublish has drawn significant attention. NCO also displays a category difference. Both the cobaltates and the cuprates require hole transport in this study: the transport properties in this study: the linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are linear Hall coefficient. While some of its tendencies are enhanced by the electronically frustrated kinetic energy operator, a phenomenon that we have previously named kinetic antiferromagnetism (KA) in the case of a single hole in the infinite $U$ limit. In this study we explicitly isolate the effect of KA by studying the $J=0$ case for various dopings.

Beginning at a doping of only a single hole in the infinitely correlated system, we track the effect of KA into the intermediate doping regime. The specific heat develops a low-temperature peak indicative of effective spin interactions, generated by a dressed hole in an AFM-spin background. The same effect is manifest in the susceptibility results where we present an $x$- and $J$-dependent effective interaction, with a small but finite AFM value in the case of $J=0$. We proceed by an explicit computation of the frequency-dependent Hall coefficient. The results show weak frequency dependence of $R_H$ and encourage the applicability of its high-frequency limit $R_H$ to practical situations. While small in magnitude compared to the spin susceptibility, we show through an identity in terms of derivatives—suggested by our numerical results—that the diamagnetic susceptibility alone may be closely connected to the temperature-dependent Hall coefficient.

This paper has two main objectives: We have recently studied the origin of the Curie-Weiss phase in Ref. 11 by...
presenting the numerical results of the $t$-$J$ model as applied to NCO. In this, the companion long paper, we provide fuller details of our results. Second, we will discuss the finite temperature $t$-$J$ model starting from the Nagaoka regime and then extend into finite dopings. The structure is as follows: In Sec. II we describe our model and the exact diagonalization technique, which we utilize for our computations. We show details of the relevant finite clusters and the inclusion of a small magnetic field perpendicular to the two-dimensional planes. Section III presents the results for the static properties: Sec. III A, chemical potential; Sec. III B, specific heat and entropy, as well as Sec. III C, the spin susceptibility. We point out the implications of electronic and spin frustration for the temperature and doping dependence of these quantities. We then investigate in Sec. III D the dynamic Hall coefficient $R_{H}(\omega)$ and compare its dc and high-frequency behavior. Last, in Sec. III E we draw a connection between the Hall coefficient and the diamagnetic susceptibility as a relation between derivatives. For all properties, we study systematically the dependence on temperature, doping, and interaction strength. In Sec. IV we conclude and summarize our results.

II. MODEL AND EXACT DIAGONALIZATION

As a model for hole doped strongly interacting Mott insulators, the $t$-$J$ Hamiltonian can be applied to the experimental situation of electron-doped NCO after an electron-hole transformation, making use of the symmetry of the Hubbard model in respect to half filling. The replacement rules are $t \rightarrow -t$, $\delta = |1 - n| = x$, and $q_{c} \rightarrow -q_{c}$, where $\delta(x)$ always refers to hole (electron) doping away from half filling and $n$ is the electron density per site. This leads to the $t$-$J$ model

$$\hat{H} = -t \sum_{\langle i,j \rangle, \sigma} \hat{P}_{G} c_{i, \sigma}^{\dagger} c_{j, \sigma}^{\dagger} \hat{P}_{G} + J \sum_{\langle i,j \rangle} \left( \hat{S}_{i} \cdot \hat{S}_{j} - \frac{\hat{n}_{i} \hat{n}_{j}}{4} \right),$$

(1)

where $c_{i, \sigma}^{\dagger}$ ($c_{i, \sigma}$) creates (annihilates) an electron of spin $\sigma$, $\hat{S}_{i}$ is the three-component spin operator, $\hat{n}_{i}$ is the number operator and $i$ specifies the lattice site. $\hat{P}_{G}$ denotes the Gutzwiller projector and the summation is over all nearest-neighbor pairs $\langle i,j \rangle$.

In the following, whenever doping $x$ is mentioned this refers to electron doping in the NCO system and hole doping in the model Hamiltonian. To compute the complete spectrum—as required for thermodynamic properties—we employ the canonical ensemble exact diagonalization technique for small clusters. The computational demand limits the analysis to quite small systems and finite-size effects are challenging to overcome. Nonetheless, careful comparison between several systems of different geometry enables us to extract the stable behavior and somewhat isolate finite-size effects. Conversely, finite-size effects can help identify the favored eigenstate of a system.

We have employed several toroidal and ladder geometries with $L=9$, 12, and 18 sites, as well as the highly symmetric icosahedral geometry. The icosahedron—a 12-site platonic solid with 20 equilateral triangular faces—shares the property of geometric frustration with the infinite system while the effect of the reduced coordination number ($z=5$) appears to have only a small, quantitative effect on the computational results [Fig. 1(c)]. Our 12-site clusters are depicted in Figs. 1(a) and 1(b). A third 12-site cluster is the simple $4 \times 3$ cluster (called torus c, not pictured).

While the numerical effort of exact computations for translationally invariant systems can be significantly reduced by exploiting space-group symmetries of the Hamiltonian, these symmetries are broken upon the introduction of a magnetic field, as relevant for the evaluation of the Hall coefficient. A magnetic field is introduced by the usual Peierls substitution which modifies the hopping $t$ between sites $i$ and $j$ by

$$t \rightarrow t(A) = t \exp \left( i \frac{2\pi}{\phi_{0}} \int_{j}^{i} A \cdot ds \right),$$

(2)

where $A$ is the magnetic vector potential and $\phi_{0}$ is the flux quantum. We define the flux threading a triangular plaquette as $\alpha = \frac{\phi_{0}}{2\pi} \int_{\Delta} A \cdot ds$. In finite systems, the value of the smallest nonzero magnetic field is limited to values of $\alpha \approx \pi / l$ where $l$ is the length of a periodic loop in the system. Through a particular gauge we can achieve $l$ equal to the number of triangular faces in the cluster, this guarantees the equality of the flux values through all plaquettes. An example is given in Fig. 1(a). A similar strategy has been followed in the case of the square lattice quantum Hall effect. We complement the computations on periodic systems with ladder systems, which enable an infinitesimal flux to be chosen due to open boundary conditions in one of the spatial dimensions [Fig. 1(b)] and the Landau gauge is adequate. In the case of the icosahedron [Fig. 1(c)] the magnetic field is applied through a (fictitious) magnetic monopole in its center, creating equal magnetic fluxes through all triangular plaquettes.

III. RESULTS

A. Chemical potential

The chemical potential is evaluated as a derivative of the free energy $F$ with respect to the particle number $N$,

$$\mu(T) = \frac{\partial F(T)}{\partial N}.$$  

The derivative has to be taken as a discrete difference of $F(N)$. To take this derivative numerically, we use the central difference formula. Hence it is suitable to define

$$\mu^{\pm} = F(N+1) - F(N)$$

and

$$\mu^{-} = F(N) - F(N-1).$$

The value of $\mu$ can then be obtained from

$$\mu = \frac{(\mu^{+} + \mu^{-})}{2}.$$  

In Figs. 2(a) and 2(b) we present $\mu(T)$ at several $J$ and dopings $x$. At low temperatures $\mu(T)$ shows degenerate Fermi behavior $(\sim T^{2})$. In the strong-correlation regime ($J=0$ and $J=0.4|\delta|$) the ground-state (g.s.) internal energy has a

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FIG. 1. (Color online) Small numbers along arrows (red) denote Peierls phases in units of the flux through a single plaquette, phases on icosahedron (c) not shown for clarity. Dotted parallelograms (black) specify boundaries of finite clusters (a) and (b). All sites on corners of parallelograms are equivalent and translations along bounding vectors $\mathbf{T}_1$ and $\mathbf{T}_2$ tile infinite lattice; shaded region in (b) forbidden for ladder. Note: different coordinate systems are used for torus and ladder. For ladder system we use the Landau gauge with $A = y \hat{x}$, (a) represents only one choice (denoted torus a) for 12 sites. Another (denoted torus b) results from ladder (b) by connecting all corners through $\mathbf{T}_1$ and $\mathbf{T}_2$ (not shown). Torus c is simple $3 \times 4$ cluster (not shown).

FIG. 2. (Color online) Chemical potential $\mu(T)$ for all accessible dopings on torus b: $x = 2/12 = 0.16$ to $10/12 = 0.83$ for $J = 0$ (a), $J = 0.4|t|$ (b), and $U = 0$ (c). Data for $U = 0$ computed for infinite non-interacting system, comparison with (a)–(c) shows relatively weak effect of interactions in the doping range $x > 0.8$. (d) Squares are experimental data for $\Delta \mu(T) = \mu(T) - \mu(0)$ from Ref. 15 Shaded area is a comparison with our data for $U = 0$ for $t = -100$ K to $t = -300$ K.

maximum at intermediate dopings $x \approx 0.5$ due to the competition of increasing number of mobile particles and reduction of the available vacancies upon decrease of doping and hence the low-temperature chemical potential changes sign here. In the high-temperature regime the doping dependence of the Hilbert-space dimension determines the limit of

$$\mu(T \to \infty) \propto T \frac{d g(x)}{dx}$$

with $g(x) = (1-x) \ln[2x/(1-x)] + \ln(1/x)$ essentially the natural logarithm of the dimension of the Hilbert space. In the projected $t$-$J$ model, $g(x)$ peaks at $x = 1/3$. For $x < 1/3$ the slope at large $T$ becomes negative. In Fig. 2 the comparison of $\mu(T)$ for different interactions shows that the impact of projected fermions near half filling is to force a negative slope of $\mu$ for $x > 0$ which occurs in the noninteracting system only for $x < 0$. This is an example of the qualitative difference between a strongly interacting Fermi system and its noninteracting counterpart.

At larger $x > 0.7$ the effect of strong interactions is gradually lost and the temperature dependence resembles that of the noninteracting Fermi system [Fig. 2(c)]. For low temperatures well below the linear range of $\mu$, finite $J > 0$ generally increases the value of the chemical potential with a stronger increase for $x$ near the Mott insulator. The high-temperature behavior is independent of $J$.

In Fig. 2(d) we compare the results with direct experimental measurements of the $T$-dependent chemical potential at $x = 0.83$. Since our results in this doping range are rather insensitive to the value of $U$, we compare with our $U = 0$ results. Here, $\mu(T)$ was obtained directly from the tight-binding density of states by the implicit equation $\langle n \rangle = 2 \int_{-\infty}^{\infty} f(e, \mu) \rho(e) e \, de$ with fixed $\langle n \rangle = 1 + x$, where $f$ denotes the Fermi-Dirac distribution function and $\rho$ is the noninteracting density of states. This fit confirms the notion of NCO as a system with an unusually small Fermi temperature scale. In Fig. 2(d) we show as a shaded region the values $t = -100$ K to $t = -300$ K, photoemission results point to a value of $|t| \approx 10$ meV $\sim 118$ K, hence $|t|$ here is more than an order of magnitude smaller than in the cuprate supercon-
ductors. In the following, when making contact with experiments, we take $t = -100$ K for simplicity.

**B. Specific heat, entropy**

In Figs. 3(a) and 3(b) we present the specific heat per site, $C_v(T)/k_B = \frac{1}{L} \frac{d\langle E \rangle}{dT}$, with $\langle E \rangle$ the thermodynamic expectation value of the internal energy. We first concentrate on the case of $J=0$, corresponding to an infinite on-site interaction. Near the band insulator the temperature dependence resembles that of noninteracting fermions, computed at fixed particle density by first determining $\mu(T)$ and then the $T$ derivative of the internal energy. When the doping is reduced, the main energy scale—manifest in a peak of $C_v(T)$ at temperature $T_m(x,J)$—continuously moves to smaller temperatures, away from the tight-binding peak. Even at our lowest doping $x=1/L$ the peak feature persists (called the KA peak). This feature is magnified in Fig. 4 for several different systems.

Numerical work on the g.s. of the problem of a single hole in an AFM spin background suggests this system to exhibit a weak effective AFM interaction $J_{eff} \approx 1/L$ due to the projected kinetic energy term only. While for one hole this excitation decays in the thermodynamic limit, $J_{eff}$ nonetheless induces a Néel ordered AFM g.s. and low-energy excitations similar to those observed in the HAF.

The validity of our computations for the case of a single hole is underscored by the entropy. In Fig. 5, the temperature dependence of the entropy shows that the low-energy features observed in the specific heat are necessary for the entropy to obtain its (known) high-temperature limit. For $T/|t| \approx 0.075$ half the entropy is recovered $[s(T)/s(\infty)=0.5]$, indicative of entropy enhancement due to electronic frustration. For intermediate dopings, the entropy reaches half its maximum at $T/|t| \approx 0.75$ and for dopings $x=0.7$—relevant to the Curie-Weiss phase of NCO—we find $T/|t| \approx 0.78$.

Within a high-temperature expansion a numerical argument was given for an S-shaped low-temperature behavior of

![FIG. 3.](image3.png)  (Color online) Specific heat per site as function of $T$ for varying $x$, computed on 18-site ($x=0.72$) and 12-site cluster ($x<0.72$), (a) $J=0$, (b) $J=0.4|t|$. Dotted lines show $C_v(T)$ for non-interacting fermions and solid line for exact results. Note: scale changes in plots of different dopings, long temperature exponential behavior at large $x$ due to finite-system induced gap in energy spectrum. Results for $x=0.72$ appeared in Ref. 11.

![FIG. 4.](image4.png)  (Color online) $C_v(T)$ at $x=1/L$ for several sets of boundary conditions, peak in specific heat is system-independent property.
we found\(\text{situation of a single hole}\)\(\text{results, we neglect the contributions at very low heat. When extracting this quantity from our numerical responding to the low-energy electronic part of the specific purely from the finite system induced gap in the spectrum and the first excited state noninteracting tight-binding electrons}C\text{markable here that applicable to a metal from Fermi-liquid arguments. It is remarka}\begin{equation}\end{equation}\text{able to consider the low-energy excitations, notably even for larger dopings than the one applicable to the Nagaoka situation.}

We now distinguish the effect of \(J > 0\) [Fig. 3(b)]. This scenario has been investigated by several authors for the case of the hole doped square lattice and the HAF.\(17,18\) For the situation of a single hole \(J > 0\) shifts the KA peak to larger temperatures \(T_m = J/t\). In Fig. 8(b) we present the peak positions as function of doping and \(J\). We find \(T_m(x, J)\) to increase monotonically both with \(x\) and \(J\) and the slope as function of \(J\) for fixed \(x\) to decrease for larger values of \(x\). Extrapolating these results at a given value of \(J\) to \(x = 0\) we obtain roughly \(T_m(x = 0) \approx J\). Our result should be distinguished from investigations on the \(t > 0\) square lattice,\(17\) where \(T_m\) was found to follow the opposite trend decreasing slightly with \(x\) at small dopings, along with a rapid suppression of the peak. This supports the notion of electronic frustration contributing to AFM correlations in the present study in opposition to competing AFM exchange coupling and the FM Nagaoka mechanism in the case of \(t > 0\), capable of quickly eliminating the low-energy spin excitations.

An experimentally interesting quantity related to the specific heat is \(\gamma\) defined by \(C_v(T) \sim \gamma T\) or \(\gamma = ds/dT\) corresponding to the low-energy electronic part of the specific heat. When extracting this quantity from our numerical results, we neglect the contributions at very low \(T\) arising purely from the finite system induced gap in the spectrum [scaling as \(\exp(-\Delta/T)\) where \(\Delta\) is the gap between the g.s. and the first excited state]. For the large doping case \(x = 0.7\) we found\(11\) \(\gamma = 45(\pm 5)\) mJ/mol K\(^2\) (\(\gamma_0 = 33\) mJ/mol K\(^2\) for noninteracting tight-binding electrons), indicating a significant many-body enhancement. Experimental results\(19\) indicate \(\gamma = 26\) mJ/mol K\(^2\). On the other hand, in the low-doping regime the linear increase with \(T\) is only observed in a very small temperature range and this range is possibly better classified as a non-Fermi-liquid regime. Roughly, \(\gamma(x) / \gamma_0(x) \approx 1\) for \(x \approx 0\). The increase in \(\gamma\) with decreased doping is also demonstrated clearly by the low \(T\) slope of \(s(T)\) in Fig. 5.

![FIG. 5. (Color online) Entropy per site for three different dopings at \(J=0\) \((x=1/12=0.08, x=5/12=0.42, x=13/18=0.72)\), horizontal lines indicate high-temperature limit \(s(T \to \infty) = k_B \ln(g(x))/N, x=0.08\) corresponds to Fig. 4. Exponential increase at low \(T\) for intermediate \(x\) due to finite system-size gap \(\Delta\).](image)

![FIG. 6. (Color online) Spin susceptibility per site \(\chi(T)\) for all accessible dopings \(x\) on 12-site torus. Dependence on \(J/t\) \(=(0, 0.1, 0.2, 0.3, 0.4, 0.5)\), indicated by arrow, blue dotted line shows bare susceptibility for noninteracting fermions of density \(n = 1 + x\) and black dotted line indicates susceptibility of isolated spin-\(1/2\) fermions of density \(n = 1\). Plots for \(x > 0.58\) appeared in Ref. 11.](image)

### C. Spin susceptibility

To gain further insight into the role of spin fluctuations and to address the existence of the experimentally observed Curie-Weiss phase of NCO,\(20\) we compute the spin susceptibility \(\chi(T)\) as function of temperature from the Gibbs free energy \(G(T, M)\), here \(M = \Sigma_{j} S_{j}^{z}\) is the total magnetization. This quantity follows from the Helmholtz free energy \(F(T, H)\) through a Legendre transformation \(G(T, M) = F(T, M) - H \frac{df}{dM} = F(T, H) + MH\). \(H\) is the externally applied magnetic field:

\[\chi^{-1} = \frac{d^2G}{dM^2}\]

can then be evaluated numerically, by constraining the canonical ensemble to the sector of minimal \(|S_{x}^{\text{tot}}|\), and those sectors where \(S_{x}^{\text{tot}}\) is raised (lowered) by \(h\).

For low temperatures, near the band limit (\(x = 1\)) we identify weakly temperature-dependent, Pauli-like behavior of \(\chi\), reminiscent of the experimentally observed spin-density wave (SDW)-phase (Fig. 6). When \(x\) is lowered past \(x = 0.75\) we find\(11\) a crossover to a Curie-Weiss-like behavior with appreciable renormalizations of the g.s. susceptibility compared to its bare value, mirroring the experimental findings. This behavior can roughly be understood by picturing electrons distributed on an empty lattice: Initially, the many-body eigenstate enables the electrons to move around without interacting directly with their neighbors, yielding a susceptibility that is \(J\) dependent only to second order. When the
density is increased ($n > 0.25$ or equivalently $n < 1.75$), electrons are forced to interact through the exchange-coupling directly and hence yield a strongly $J$-dependent magnetic susceptibility. The emergence of the Curie-Weiss phase is further supported by the strong magnetic-field suppression of the Heikes-Mott term in the thermopower within the $t$-$J$ model, an effect due to spin entropy$^5$ as was demonstrated in Ref. 11.

For temperatures larger than a certain crossover temperature (which we call $T^*$), for all dopings, $\chi(T)$ can be described through the Curie-Weiss form

$$\chi(T) = \frac{1}{3} \frac{L}{V} \frac{1}{k_B(T-\theta)^2}$$

with the Weiss temperature $\theta$ and the effective magnetic moment $\mu_{\text{eff}}$, $V$ is the volume of the crystal, and $L$ is the number of sites. For all accessible system sizes—in particular torus a, b, c (Fig. 1)—we compute $\chi(T)$ to extract $\theta$, $\mu_{\text{eff}}$, and $T^*$, the parameters characterizing the magnetic properties at a given doping and interaction strength. To determine $\theta$ and $\mu_{\text{eff}}$, we fit the high-temperature part of our computations for $\chi(T)$ to the function $f(T)=a/(T-b)$ [compare Fig. 7(a)]. In Figs. 7(a)–7(c) we present the inverse susceptibility at several different dopings near the Mott-insulating phase. The results for $\theta$ and $\mu_{\text{eff}}$ are shown in Figs. 7(d)–7(f). For all $x$ and $J \geq 0$, $\theta$ remains negative indicating AFM correlations. $\theta$ shows a linear increase when the doping is increased or the interaction is decreased. In short, as mentioned in Ref. 11, $\theta$ can be collapsed into the form $\theta(x,J) = -c J_{\text{eff}}(x)$ where

$$J_{\text{eff}}(x) = J(1+c''x|t| + c'x|t|)$$

with $c=4.0$, $c'=0.01425$, and $c''=-0.9175$. In contrast to the relation given by Anderson et al.$^1$ in the case of the $t>0$ square lattice, in our situation of $t<0$ even at $J=0$ there is a slightly negative intercept suggesting AFM spin interactions in the low-temperature regime of the electronically frustrated $U=\infty$ Hubbard model, indicative of kinetic antiferromagnetism (KA).
On the other hand, once \( J > 0 \) is allowed, this intercept grows strongly and expresses dominant AFM correlations [Fig. 7(e)]. We compare these results with experiments for various dopings \((x > 1/2)\) of NCO. Among others, the cases of \( x = 0.75, 0.82, 0.84 \) and \( x = 0.6, 0.7, 0.8 \) and 0.9
diagram is set by the interaction strength. As was shown
undergoes a transition from an AFM-ordered to a paramag-
etically investigated. The comparison with experiment
proves difficult due to the large number of structural
phases, \(^{21}\) the role played by the Na ions in the crystal, and
the large quantitative and qualitative difference between
powder and single-crystal samples, \(^{23}\) However, the Weiss
temperature appears to lie between 100 and 200 K near
\( x = 0.7 \) [varying considerably with \( H \mid c \) and \( H \mid a \) (Ref. 23)]
and was measured to be smaller at larger dopings. \(^{21}\) Hence at
least for \( x > 0.5 \) the \( t-J \) model description yields reasonable
experimental agreement.

The effective magnetic moment \( p_{\text{eff}} \) in units of \( \mu_B \) has a
value close to that obtained for localized spin-\( 1/2 \) particles at
half filling \( \mu_{1/2} = g \times \gamma(s+1) = \sqrt{3} \) and decays linearly for
\( x \rightarrow 1 \).

The inverse susceptibility shows a minimum at a tempera-
ture \( T^* \) which becomes more pronounced with increasing \( J \).
At low temperatures the system prefers antiferromagnetic
order which is \textit{strengthened} by \( J > 0 \). Thermal spin fluctuations
break this order and when \( T \) is increased to values of order \( J \)
a small perturbative \( B \) field is sufficient to align spins anti-
parallel to the field. Hence the susceptibility is largest at this
temperature. When \( T \) is further increased, the ability of the
field to flip spins is weakened by thermal fluctuations, con-
sequently, \( \chi(T) \) decreases. The behavior of this peak as func-
tion of hole doping away from the Mott insulator has already
been investigated within a high-temperature expansion. \(^{16}\)

Due to the small size of the systems available to our
study, it is difficult to confirm the results for the doping
dependence. \(^{16}\) Here, we study the behavior as function of \( J \),
considering several dopings near half filling and several
system geometries [Fig. 8(a)]. The figure indicates that \( T^*/|t| \)
roughly scales as \( J/|t| \) hence the scale at which the system
undergoes a transition from an AFM-ordered to a paramag-
netic state is set by the interaction strength. As was shown
for the case of \( J = 0 \) and \( t > 0 \) in the hole doped system, \(^{2}\) a
single hole favors AFM Néel order and numerical results \(^{9}\)
indicate that the triangular lattice Heisenberg model also fa-
vors Néel order—at least in the g.s. and the elementary
excitations—hence it is not surprising that a finite \( J > 0 \)
emphasizes this tendency. This is in opposition to the case of
\( t < 0 \) where the kinetic energy term favors ferromagnetism
and competes with the Heisenberg interaction preferring
AFM.

D. Hall coefficient

Experimentally, NCO has been shown to exhibit unusual
thermoelectric properties, resulting from strong electron cor-
relations. Wang \textit{et al.} \(^{6}\) measured the dc-Hall coefficient as a
function of temperature at several dopings showing unex-
pected temperature dependence with an oscillatory behavior
below 200 K and a rapid linear increase for temperatures
above 200 K.

\begin{equation}
R_{H}'(T) = \lim_{\omega \to \infty} R_H(T, \omega) = \lim_{B \to 0} \left( -\frac{ivL(J_x, J_y)}{Be^2 \left( \tau_{c, e} \right)^2} \right).
\end{equation}

Here, \( e \) is the electron charge, \( J_x \) (\( J_y \)) are the currents in \( x \)
(\( y \)) directions, \( v \) is the unit-cell volume, and \( \tau_{c, e} \) is the diag-

FIG. 8. (Color online) (a) AFM-paramagnetic crossover tempera-
ture as function of \( J \) for several dopings on 12-site torus clus-
ters a, b, c. (b) Evolution of \( T_m \) as function of \( J \), values averaged
over three 12-site clusters a, b, c.

Much theoretical work has been invested into the study of
\( R_{H} \) on square lattices \(^{3,12,27-39} \) both for g.s. properties and for
finite temperatures. For the triangular lattice, it was possible
to predict the high-temperature behavior within a high-
temperature expansion of the \( t-J \) model \(^{13,12} \) in its high-
frequency limit. Several authors have also attempted to de-
scribe this behavior through Boltzmann transport theory \(^{36,37} \)
within the relaxation time approximation, where \( R_{H} \) is ex-
pressed as an integral over the Fermi-surface curvature and
the relaxation time \( \tau_i \) is taken to be isotropic. For large
dopings—\( x > 0.7 \)—where the holelike Fermi surface of NCO
approximates a circle, this leads to the same high-
temperature linear coefficient as the \( t-J \)-high-temperature ex-
pansion, however, the behavior completely diverges in the
dense regime (\( x < 0.7 \)), possibly due to the misleading re-
placement of the interacting Fermi surface by its noninteract-
 ing counterpart. Also, at low temperatures—where the relax-
ation time is anisotropic—the oscillatory behavior seen in
experiments is not reproduced.

To shed some light on the low-temperature and strongly
correlated behavior, we start from the high-frequency limit \(^{4}\)
for \( R_{H}'(T, \omega) \):

\begin{equation}
R_{H}'(T, \omega) = \lim_{\omega \to \infty} R_{H}(T, \omega) = \lim_{B \to 0} \left( -\frac{ivL(J_x, J_y)}{Be^2 \left( \tau_{c, e} \right)^2} \right).
\end{equation}
onal part of the stress tensor. To ensure the effect of strong correlations within this limit, it is necessary to force the hierarchy $t < \omega \ll U$. We enable this by projecting out double occupancy using the Gutzwiller projector, hence the $t-J$ model is the appropriate description.

Our plan is as follows: we show the connection of this high-frequency limit—not measurable in experiments—to the dc or low-frequency behavior. Then we evaluate the dependence on the parameters $T$, $J$, and $x$ for several systems. Hence for the first part we compare $R_H^T$ with the expression derived from the Kubo formulas for the electrical conductivities

$$
\sigma_{\alpha,\beta} = \frac{ie^2}{H_1} \left( \tau_{\alpha,\beta} \right) - \frac{1}{Z} \sum_{\mu,\nu} \frac{\exp - (\beta \epsilon_\mu) - \exp - (\beta \epsilon_\nu)}{\epsilon_\mu - \epsilon_\nu - \omega - i \eta} \langle v [J_\beta, \mu] \rangle \times \langle \mu | J_\beta | \nu \rangle ,
$$

with $\epsilon_\nu$ the $\nu$th energy eigenvalue of $\hat{H}$ and $Z$ the canonical partition function. In terms of this expression the Hall coefficient is given by

$$
R_H^T(\omega) = - \frac{\partial}{\partial B} \left( \frac{\sigma_{x,y}(\omega)}{\sigma_{x,x}(\omega)} \right) \bigg|_{B=0} .
$$

As a transport object $R_H$ is a demanding quantity to study on finite clusters. The effect of boundary conditions and their finite-size implications are difficult to eliminate. We introduce a small magnetic field through the Peierls substitution [Eq. (2)] which implies only on the orbital motion of the electrons and does not couple to the spin. Studying $R_H$ on toroidal and spherical geometries reduces the finite-size effects on the transport, however, these periodic systems constrain the smallest accessible flux $\phi_0$ to $\pi/L$. For $L=12$ this is already a considerable flux. An alternative are ladder systems such as in Fig. 1(b) where the limit $\phi_0 \rightarrow 0$ is possible. However, ladders have additional finite-size effects impacting on the transport quantities.

For several dopings ($x=0.08, 0.67, 0.75, 0.83$) we compute $R_H^T(\omega)$ as a function of temperature both on the torus and the icosahedron. To take into account the finite size of the system, a broadening $\eta$ of the energy equal to the mean level spacing of the current matrix elements was employed, this requires $\eta = 3|t|$. In Fig. 9 we show $R_H$ as a function of temperature at three different frequencies. Comparing $\omega=0$ to $\omega=\infty$ we find that the dc limit is qualitatively the same as the high-frequency limit, however, the overall temperature dependence is scaled by a renormalization factor $s = R_H^T(T, \omega=0)/R_H^T(T, \infty)$ where $s$ is only weakly temperature dependent. For the torus, $s$ was found to be 0.83 (0.78) at $x=0.83$ ($x=0.75$). At $x=0.083$ the many-body renormalization is stronger.

Conversely, in Fig. 10 we show the frequency dependence of $R_H$ at several temperatures and different dopings. Similar to the work done by Grayson et al. on the Hall angle, finite frequency experiments may be helpful to confirm the weak frequency dependence seen in our computations: With $|t|=10$ meV results in the frequency range $\omega \approx 3|t|=30$ meV could be compared to our theoretical results and hence lend experimental support for the usefulness of the high-frequency limit of $R_H$.

We have computed two additional quantities related to the dc limit as checks of the validity of the dc computations. Zotos et al. derived an expression for the g.s. expectation value, $R_H^T(T=0) = |J^0| \ln D(n)$ with $D$ the charge stiffness $D = \frac{d^2 \langle E_0 \rangle}{d \phi^2}$, $\langle E_0 \rangle$ the g.s. energy, and $\phi$ a constant flux inducing a persistent current. In this fashion, we evaluated the doping dependence of the g.s. and found $R_H^T > 0$ in the experimental regime near $x \approx 0.7$ and a divergence $R_H^T \rightarrow -\infty$ as the Mott limit is approached. The second check refers back to the basic definition of the dc-Hall coefficient $R_H = -E_x/(Bj_x)$ and computing this directly on a ladder system. This computation showed no change of sign for $x=0.7$ as a function of $T$ and a qualitatively similar behavior to the $\omega=\infty$ result.

![FIG. 9. (Color online) $R_H^T(\omega, T)$ on 12-site torus at several frequencies for several dopings and $J=0$.](image)

![FIG. 10. (Color online) $R_H^T(\omega)$ at several temperatures and dopings as function of frequency on 12-site torus a for $J=0$. Temperatures in all plots $T/|t|=(0.5, 1, 2, 4)$.](image)
Encouraged by the notion of weak frequency dependence, we studied $R_H = R_H(\omega = \infty)$. We have studied several 12 site tori, several doping values on an 18 site torus, the icosahedron and several ladders.

The g.s. $R_H(x)$ shows a Drude dropoff near the band insulator and a saddle point around $x=0.5$ as was reported by the authors in Ref. 11. When the strong-correlation regime near the Mott insulator is approached $R_H(x)$ moves from $T=|t|$ to $T=1.1|t|$ while the minimum in $R_H$ shifts from $T=0.75|t|$ to $T=1.1|t|$. At $x=0.67$ the behavior around $T=|t|$ is similar, however, an additional peak in the specific heat emerges at $T<0.5|t|$. A qualitatively similar feature is seen in other clusters, however, its magnitude is dependent on the system geometry. Hence we do not attempt to extract quantitative information in the $T$ range below $T=0.5|t|$. Nonetheless, it is important to note that with decreasing doping a second energy scale—caused by spin interactions—becomes dominant. At small doping $x=0.16$ and $x=0.08$ the specific heat becomes strongly $J$ dependent and the main peak in the specific heat—as discussed in the first section—increases significantly with $J$. The variations in $R_H(T)$ with $J$ are also more pronounced.

![Figure 11](https://example.com/figure11.png)

**FIG. 11.** (Color online) (left) $R_H(T)$ for several $x$ and $J$ for torus b. (right) $C_v(T)$ for several $x$ and $J$ on same torus.
The exact temperature dependence of the onset of the linear increase for $t_{e}=100$ K, in the lower plot our numerical results are scaled to match the slope of the linear part at $T>200$ K in the experimental results.

Assuming $t=-100$ K we compare our results to the experimentally available data at $x=0.31$ and $x=0.71$, this was done in Ref. 11 for $x=0.71$. In Fig. 12 the comparison at $x=0.31$ shows good agreement for the slope at high $T$ and reproduces the temperature scale for the minimum near 25 K. For $x=0.71$ the temperature scale for the minimum near 100 K is seen both in experiment and theory. The slope of the high-temperature limit differs by a factor of 4 and was adjusted in the plot for better comparison. Also, the theory does not show the change of sign as function of temperature. However, the oscillatory behavior below $T=100$ K and the onset of the linear increase for $T>200$ K agrees well with experiment. Hence the exact temperature dependence of $R_H$ remains a challenging task—both on the theoretical and experimental side.

E. Diamagnetic susceptibility

Last, we study the orbital susceptibility per site,

$$\chi_d(T) = \frac{1}{N} \frac{\partial^2 F}{\partial B^2},$$

where $F$ is the free energy and $B$ is the magnitude of the perpendicular magnetic field. We define the dimensionless flux $\alpha = BA/\phi_0$ through a triangular plaquette with $A=\sqrt{3}a^2/4$ the area of the plaquette and $a$ the lattice spacing. We get

$$\chi_d = \frac{1}{L} \frac{\partial^2 F}{\partial \alpha^2} \left( \frac{\partial \alpha}{\partial B} \right)^2$$

It is useful to employ ladder systems as shown in Fig. 1(b). Defining $t=\hbar^2/2ml^2$ we have

$$\frac{1}{L} \chi_d = -\frac{2m}{\hbar^2} A^2 \mu_B^2 \frac{1}{L} \frac{\partial f}{\partial \alpha},$$

where $f$ is the dimensionless $F/t$ and $\mu_B$ is the Bohr magneton. In Fig. 13(a) we present the diamagnetic susceptibility and its first two derivatives with respect to $T$. For free electrons in an infinite system, $\chi_d$ corresponds to electrons moving in circular orbits with radii inversely proportional to the magnetic field. $R_H$ measures the polarization induced in a system with a stationary current and perpendicular magnetic field. It is hence reasonable to ask for a connection between these two quantities. Our numerics suggest the relation

$$T \frac{\partial^2 \chi_d}{\partial T^2} = c(x) \frac{\partial^2 R_H}{\partial t^2},$$

where $c(x)$ is a function depending on doping $x$.

In Fig. 13(b) we display our numerical result for $R_H(T)$ at a doping relevant to the experimental situation compared to the result obtained from integrating Eq. (6) and choosing the two integration constants appropriately. For $J=0$ this was done by the authors in Ref. 11. Ignoring the range for $T<0.5|t|$ which may be strongly influenced by finite-size effects, we find that both curves show a minimum at $T=|t|$.
In our case antiferromagnetism is effective spin interactions regime with a low entropy recovery temperature. The effect of electronic frustration is at very low are manifest in a peak structure in the specific heat emerging for intermediate dopings. The effect of electronic frustration is further supported by entropy enhancement in the underdoped regime with a low entropy recovery temperature.

We have studied both the diamagnetic and spin susceptibility. For the spin susceptibility we extract a relation for effective spin interactions \( \theta(x, J) \) valid for all dopings. This stands in contrast to Anderson’s formula for FM \( t > 0 \), since in our case antiferromagnetism is supported rather than suppressed by doping. In the experimentally relevant doping range of \( x > 0.5 \) we offer an explanation for the crossover between the Curie-Weiss and SDW metallic phases, as observed in experiments. For the diamagnetic susceptibility \( \chi_d \) a connection is obtained between \( \chi_d \) and the temperature-dependent Hall coefficient.

The dynamic Hall coefficient reveals a weak frequency dependence and a qualitatively similar dc and high-frequency behavior, with stronger renormalizations close to the Mott limit. This strengthens the practical relevance of the high-frequency limit \( R_{Hd} \). The temperature dependence confirms the linear slope at high temperatures and reproduces the experimentally observed minimum near \( T = 100 \, \text{K} \) at \( x = 0.71 \).

In conclusion, the \( t-J \) model is applicable to NCO for \( x > 0.5 \) and shows that the interface of strong correlations and metallic behavior indeed describe the experimental situation here. For \( x < 0.5 \) the frustrated \( t-J \) model leads to subtle effects of strong correlations, even in the case of \( J = 0 \). Our results show that for the proper description of experimentally observed normal-state properties there—such as the charge-ordered insulator at \( x = 0.5 \) or the paramagnetic metal for \( x < 0.5 \)—an extension of the simple \( t-J \) model would be necessary.

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14. In numerical results with g.s. degeneracies this behavior is somewhat obscured, this is a finite-size effect.
20. This discussion expands and supplements the results in Ref. 11 obtained by the same authors.