Charge instabilities at the metamagnetic transition of itinerant electron systems

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We investigate instabilities in the charge channel in the vicinity of (meta)magnetic transitions of itinerant electron systems. Based on a weak-coupling analysis we argue that in a one-band t-t' Hubbard model near the van Hove filling and dominant ferromagnetic fluctuations it is difficult to account for a microscopic mechanism for a d-wave Pomeranchuk deformation of the Fermi surface. A similar deformation has been considered for the metamagnetic transition in $Sr_3Ru_2O_7$. As an alternative we discuss the possibility of charge inhomogeneity on the nanoscale. This extends the analogy of the metamagnetic transition to a liquid-gas transition.

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I. INTRODUCTION

Over the last decade, the perovskite strontium ruthenates have become a model system for the study of correlated electrons. While the infinite-layer compound $SrRuO_3$ is an itinerant ferromagnet[1] with Curie temperature of $T_C=165$ K, single-layer Sr_2RuO_4 is a triplet superconductor[2] below $T_c=1.5$ K. Above T_c it is a well-studied strongly anisotropic Fermi liquid.[3] By substituting Sr with La and thereby enhancing the Fermi surface (FS) volumes, it can be brought into the vicinity of ferromagnetism (FM).[4] Double-layered $Sr_3Ru_2O_7$ interpolates between these two limiting cases in an exciting way. While first experiments found FM, subsequent research[5] made clear that the magnetization M is zero in the absence of a magnetic field B. However at a direction-dependent critical field strength B_m between 4 and 8 T, $M(B)$ shows a steep increase below $T\sim 2$ K, referred to as a metamagnetic transition.[6,7] Around B_m, there are indications of non-Fermi-liquid behavior—e.g., in the specific heat and the T dependence of the in-plane resistivity.[6,7] Theoretically, the metamagnetic transition with a single jump in the magnetization can be understood in a Stoner-type model with a FS near a van Hove singularity in the density of states.[8,9] Indeed a number of experiments[3] for the single-layer system Sr_2RuO_4 and band structure calculations[10] for both Sr_2RuO_4 and $Sr_3Ru_2O_7$ confirm the presence of a nearby van Hove singularity for the FS derived from the Ru d_{xy} orbitals.

A new twist occurred when improved sample quality revealed at least two jumps[11,12] in the magnetization when B is increased. Most interestingly, in the critical field range between the two jumps of Ref. 12, the residual resistivity ρ_0 is about twice as high as below and above the jumps. As a function of decreasing temperature, $\rho(T)$ saturates already at $T=1$ K in the critical field range and the T dependence above is of non-Fermi-liquid type. The high ρ_0 can be understood if one assumes the formation of domainlike structures in the critical range.[13] Recently, two works[13,14] suggested the occurrence of a $d_{x^2-y^2}$-wave-like Fermi surface deformation (dFSD). In a one-band picture of the two-dimensional (2D) square lattice, the dFSD splits the two inequivalent van Hove points and thereby gains energy if the forward scattering in the charge channel has an attractive d-wave component. This possibility has been analyzed for Hubbard-like models by a number of authors.[15–19] An experimental realization of the effect has not been found yet. For double-layer ruthenate this picture is, however, quite appealing.[13] The occurrence of two magnetization jumps can be understood in the dFSD framework as first-order entry into and exit out of the phase with reduced FS symmetry.[14] Moreover, the sensitivity of the experimental picture with respect to sample quality seems to fit the scenario of an unconventional order parameter. We note that while the chemical-potential-tuned quantum phase transition into the dFSD state is typically first order, the thermal transition at T_c is second order over a wide parameter range in the simple forward-scattering model.[20,21]

In this work we analyze how such a scenario can be motivated in the one-band t-t' Hubbard model on a 2D square lattice. We specify the parameter region in which the dFSD scenario is favorable. Furthermore, we reinvestigate the Stoner model for the metamagnetic transition.[8] We point out that in this scenario, the magnetization jump is located in an unstable density region. This extends the analogy to a liquid-gas transition already emphasized by Green *et al.*[23] Around the transition charge inhomogeneity might occur and give rise to a higher residual resistivity. We also describe a possibility how more than one jump in the magnetization can occur in this model.

In layered strontium ruthenates there are several bands which cross the Fermi surface. Our reduction to a one-band model for both scenarios for the double-layer compound is a severe simplification. However, band structure calculations[10] and the available experimental data for $Sr_3Ru_2O_4$,[6] in particular for single-layer material,[14] are consistent with a peak in the density of states near the Fermi level. The assumption underlying our treatment is that the magnetic phenomena are caused by "active" bands which are responsible for this density of states peak. The other Fermi surfaces with innocent densities of states are assumed to be spectators. As argued later on, they could also screen charge inhomogeneities induced by the active Fermi surfaces.

II. MICROSCOPIC MECHANISM FOR d-WAVE FS DEFORMATION

So far, the microscopic origin of an attractive d-wave component in forward scattering has not been addressed in

the context of metamagnetism. In the following we analyze this question based on the one-band t-t' Hubbard model on a 2D square lattice. The Hamiltonian is given by

$$H = -t \sum_{\langle i,j\rangle \mathrm{NN}}^{s} c_{i,s}^{\dagger} c_{j,s} - t' \sum_{\langle i,j\rangle \mathrm{NNN}}^{s} c_{i,s}^{\dagger} c_{j,s} + U \sum_{i} n_{i\uparrow} n_{i\downarrow}, \quad (1)$$

where t and t' denote the hopping amplitudes between nearest- and next-nearest-neighbor sites i and j on the two-dimensional square lattice. U is the repulsive on-site interaction on the sites i. As the strontium ruthenates which we are interested in are correlated Fermi liquids, we consider the case of weak to moderate couplings and take, e.g., $U \approx 3t$.

In zero magnetic field we compute the effective forward scattering in the charge channel, $V_c(\vec{k},\vec{k}')$. It describes the scattering of quasiparticles with wave vector $\vec{k}$ and spin projection s and $\vec{k}'$, s' to $\vec{k}+\vec{q}$, s, and $\vec{k}'-\vec{q}$, s' in the limit $\vec{q} \to 0$. For the dFSD to lower the energy of the system, $V_c(\vec{k},\vec{k}')$ should contain an attractive d-wave component with respect to $\vec{k}$ and $\vec{k}'$—e.g.,

$$V_c(\vec{k},\vec{k}') = -V_{c,2}(\cos k_x - \cos k_y)(\cos k_x' - \cos k_y') + V_r, \quad (2)$$

with $V_{c,2} > 0$ and some rest V_r. We now describe that for our model and dominant ferromagnetic tendencies—as observed in the layered ruthenates—the effective charge forward scattering at low T does not contain an attractive d-wave component and hence does not support the conjectured dFSD.

A perturbative but unbiased way to arrive at effective low-energy or low-T interactions is the functional renormalization group (FRG). In fact, the dFSD or Pomeranchuk tendencies in the 2D Hubbard model were revealed using the FRG.[16] Here, we use the temperature-flow FRG.[23] It has the additional advantage of not being blind with respect to long-wavelength particle-hole fluctuations. Using this method, a low-T p-wave superconducting instability was found for a FS similar to that of the γ band in Sr_2RuO_4.[23] A second-order transition into a state with broken symmetry is indicated by a divergence of the corresponding susceptibility at T_c when T is lowered.

The FRG flow is started at high temperatures of the order of the bandwidth with pure on-site repulsion U. In the flow to lower T, one-loop corrections summed by the FRG flow generate a detailed dependence of the interactions $V_T(\vec{k}_1,\vec{k}_2,\vec{k}_3)$ on the incoming wave vectors $\vec{k}_1$, $\vec{k}_2$, and the outgoing $\vec{k}_3$. In the Hubbard model with $U=3t$ near the van Hove filling, the FRG basically finds two regimes.[23] One is for $|t'| < |t|/3$, where the leading ordering tendencies are either in the antiferromagnetic channel for strong nesting or, if the nesting is weaker, in the $d_{x^2-y^2}$-wave pairing channel.[23] For $|t|/3 < |t'| < 1/2$, the leading instability is in the ferromagnetic channel. With bare on-site interactions the charge channel never becomes leading. However, e.g., the forward scattering $V_{c,T}(\vec{k},\vec{k}')$ can develop a pronounced dependence on $\vec{k}$ and $\vec{k}'$. This can give rise to subdominant instabilities such as FS deformations.[16] In Fig. 1(a) we show the FRG result for $V_c(\vec{k},\vec{k}')$ at low temperatures above the runaway flow of the leading tendency, obtained with the FRG scheme of Ref. 23. The dashed line is for temperatures above a d-wave pairing instability at the van Hove filling for $t'=0.25t$ and $T=0.09t$. One can clearly observe the attractive d-wave component in $V_c(\vec{k},\vec{k}') \approx -\cos 2\theta_{\vec{k}} \cos 2\theta_{\vec{k}'} + V_r$ when $\theta_{\vec{k}}$ moves around the FS with $\theta_{\vec{k}'}$ fixed at $-\pi[\vec{k}' \approx (-\pi,0)]$. The solid line is near van Hove filling and $t'=0.44t$ in the ferromagnetic regime at $T=0.04t$. Now the component $\propto \cos 2\theta_{\vec{k}}$ is positive; i.e., there is no sign for an attraction in the d-wave channel. The coupling to external static and uniform sources in this channel does not increase for $T \to 0$ [Fig. 1(b)].

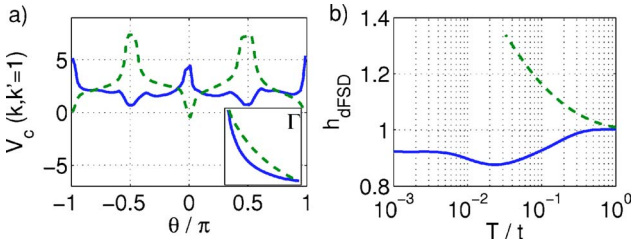

FIG. 1. (Color online) (a) Effective forward scattering in the charge channel $V_c(\vec{k},\vec{k}')$ with $\vec{k}'$ fixed near $(-\pi,0)$ $(\theta=-\pi)$ obtained with a 96-patch T-flow FRG. The inset shows the Fermi surfaces. (b) T flow of the coupling to static external dFSD fields, averaged around the FS. In all plots, the solid lines are for $t'=0.44t$, the dashed lines for $t'=0.25t$.

These results reflect the influence of the spin fluctuations on the forward scattering. Quite generally, the spin fluctuations renormalize the charge forward scattering between the parts of the FS connected by the dominant spin fluctuation wave vector $\vec{q}$ to stronger repulsion. In the antiferromagnetic (AF) and d-wave pairing regime the strong (π,π) fluctuations between the van Hove points create the + lobe of the d wave in $V_c(\vec{k},\vec{k}')$ for small t'. The attraction for $\vec{k} \approx \vec{k}'$ is a precursor of the pairing tendencies. In the ferromagnetic regime, the dominant spin fluctuations have $\vec{q} \approx 0$—i.e., make $\vec{k} \approx \vec{k}'$ more repulsive. Hence the d-wave charge forward scattering turns out to be slightly repulsive rather than attractive. In fact, there is not any clearly attractive component in $V_c(\vec{k},\vec{k}')$ in the ferromagnetic regime. When we move the FS farther away from the van Hove points by a density change or by a weak Zeeman splitting, the ferromagnetic instability is cut off. Nevertheless, the main features of the charge forward scattering remain robust.

We conclude that at least in a single-band scenario with dominant ferromagnetic fluctuations, it is difficult to identify a microscopic mechanism for a d-wave FS deformation. Conversely, if we assume that the relevant FS of $Sr_3Ru_2O_7$ is on the AF–d-wave side, the spin response should be strongest near the wave vector $\vec{q}=(\pi,\pi)$ connecting the two van Hove regions. This seems unlikely in view of the strong ferromagnetic tendencies in all layered strontium ruthenates, and in inelastic neutron scattering there is no sign of these fluctuations.[21] Furthermore, at least in the Hubbard model, the dFSD tendencies are always weaker than other instabilities. Hence, we would need a mechanism that suppresses the leading tendencies.

III. MICROSCOPIC PHASE SEPARATION SCENARIO

As we have seen, the *d*FSD scenario is difficult to motivate starting from a two-dimensional microscopic model with predominant ferromagnetic correlations. Therefore we now turn to a second scenario for the metamagnetic transition that exploits the vicinity to ferromagnetism. This ferromagnetic tendency is common to all strontium ruthenates mentioned in the Introduction. In contrast with the *d*FSD instability it fits readily into the picture drawn by a FRG treatment, as FM is found to be the leading instability of the weakly coupled Hubbard model near the van Hove filling for $1/3<|t'|<1/2$.[24] At the van Hove filling, the density of states diverges logarithmically. For this situation, the metamagnetic transition was modeled by Binz and Sigrist[8] using a mean-field theory with a density of states,

$$\rho(\epsilon) = \frac{1}{2W} \ln[W/|\epsilon - \epsilon_{\mathrm{VH}}|], \tag{3}$$

in a band $-W<\epsilon-\epsilon_{\mathrm{VH}}<W$. The total density n and the distance to the van Hove filling at $n=1$ can be adjusted by ϵ_{VH}. We define a magnetic field h which shifts the band energy of spin-up electrons by $+h$ and by $-h$ for the spin-down electrons. Setting $k_B=1$, the grand potential is given by

$$\Omega_0(\mu,h,T) = -T\sum_{s=\pm} \int d\epsilon \rho(\epsilon) \ln[1+e^{-(\epsilon-\mu-sh)/T}]. \tag{4}$$

The electrons interact with an on-site interaction $U>0$. This term is treated in mean-field fashion as an exchange potential[25] $-U(n_\uparrow-n_\downarrow)/4$ with average densities $n_\uparrow$ and $n_\downarrow$. For adding this to the band energy it is convenient to go over to a thermodynamic potential which depends on the magnetization $m=n_\uparrow-n_\downarrow$. The noninteracting free energy density $f_0(n,m,T)$ of the electrons with density $n=n_\uparrow+n_\downarrow$ and magnetization m is found as

$$f_0(n,m,T) = \Omega_0(\mu,h,T) + \mu n + hm. \tag{5}$$

So the interacting free energy reads

$$f(n,m,T) = f_0(n,m,T) + Un_\uparrow n_\downarrow. \tag{6}$$

Minimizing the Gibbs potential

$$g(T,h,n) = f(n,m,T) - hm \tag{7}$$

with respect to m yields the magnetization versus h and T. Binz and Sigrist[8] found a density region near the van Hove filling at low T where m exhibits a sudden jump at a critical magnetic field h_m. Coming down from high T at h_m, m increases strongly and finally saturates below a temperature scale T_m. Impurities cut off the logarithmic growth of the density of states (DOS) near the van Hove filling. Hence, they are expected to smear out the metamagnetic transition. In this simple model, the magnetization does not depend on the direction of the h field, but this effect can be taken into account, e.g., by including anisotropic interactions.

In Fig. 2(a) we show the steplike increase of the magnetization m with increasing magnetic field h for different electron densities n, equivalent to the data of Ref. 8. In Fig. 2(b) we show the derivative of the Gibbs potential density

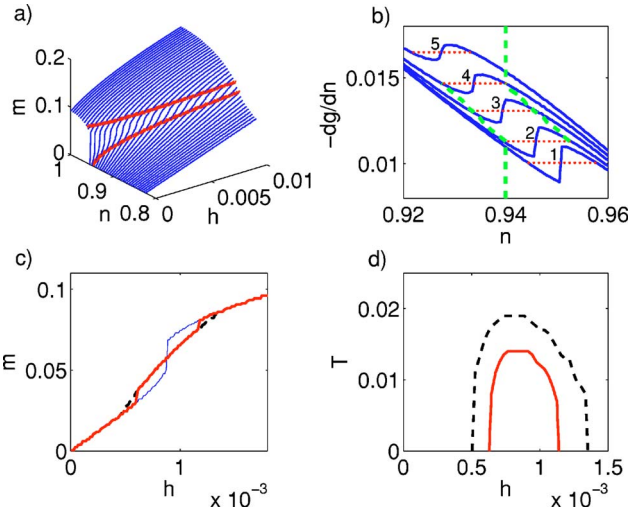

FIG. 2. (Color online) (a) Magnetization m at temperature $T=0$ vs magnetic field h in units of the half bandwidth W and density n, $U=0.4W$. The two thick lines bound the phase-separated region of the Maxwell construction. (b) $-\mu=-\partial g/\partial n$ for $h=0.0002-0.0019W$ with h increasing from curve 1 to 5. The dotted lines indicate the Maxwell construction and span the phase-separated region in the upper left plot. The dashed lines show the density components vs h for average density $n=0.94$. (c) m at $T=0$ vs h for total density $n=0.94$. The thin line is for the homogeneous case; the dashed line is the Maxwell construction allowing for zero interface energy $g_I=0$. The thicker line with two jumps is for $g_I=5\times10^{-7}W$ per unit area. (d) Boundaries of the inhomogeneity regions for total density $n=0.94$ vs h and T. The dotted lined is again for $g_I=0$ and the thick line for $g_I=5\times10^{-7}W$.

$g(T,h,n)$ with respect to n versus n for different h. For the critical h values h_m where the jump in m occurs, the curvature of $g(T,h,n)$ with respect to n, corresponding to the inverse compressibility, is negative. Hence, at the metamagnetic transition, the system is unstable with respect to phase separation. If we plot $g(T,h,n)$ versus n, we find a small upward dent in the critical density range.

In this range we can minimize the energy by the Maxwell construction—i.e., by bridging the dent from below with a straight line which lies below $g(T,h,n)$ between $n_<(h)$ and $n_>(h)$. The average density is a mixture between a less dense phase $n_<(h)$ with lower m and a denser phase $n_>(h)$ with higher m. The mixing ratio $0<p<1$ is determined by

$$pn_<(h) + (1-p)n_>(h) = n. \tag{8}$$

When we increase h from below at a fixed total density n, we enter the unstable region at a certain lower critical field $h_<(n)$ with $n_<(h_<(n))=n$. In Fig. 2(b) for $n=0.94$ this occurs for h a little higher than $h=0.0005W$ for curve 2. For curve 3 the stable solution is a mixture between $n_<(h)$ and $n_>(h)$. The path of $n_<(h)$ and $n_>(h)$ with increasing h is shown by the dashed lines. Going up, the weight p of $n_<(h)$ decreases until it vanishes for curve 4. In the coexistence region, $\mu=\partial g/\partial n$ is constant and the inverse compressibility $\kappa^{-1} \propto \partial^2 g/\partial n^2$ vanishes. With increasing h, the instability region moves to smaller densities, and above an upper critical field

$h_>(n)$, the homogeneous density $n=0.94$ has the lowest Gibbs energy again (curve 5). The wiggle in $\partial g/\partial n$—i.e., the energy difference between the phase-separated and homogeneous solutions—becomes smaller with increasing h.

Again, this is very similar to a liquid-gas transition. There the intensive tuning parameter is the temperature T instead of h. At the transition, the conjugated extensive quantity, the entropy, jumps upwards. As a function of the volume V, the Gibbs potential has negative curvature around the critical volume where the transition occurs, and the isothermal lines in the pressure- ($P=-\partial F/\partial V$) volume diagram exhibit a similar wiggle as $\mu=\partial g/\partial n$ in our case. Hence the inhomogeneous phase at the metamagnetic transition just corresponds to the coexistence regime in the liquid-gas transition. This extends the analogy already emphasized by Green *et al.*[22]

In our model the density difference between the two components of the mixture is rather weak of the order 0.01/site [see Fig. 2(b)]. In a more realistic model including long-range Coulomb interactions, the phase separation will be frustrated on a microscopic length scale.[26] Then the two-phase mixture will create a pattern of nanodomains. Stripe-like domains are likely candidates. Here we speculate that this inhomogeneity may be responsible for the observed plateau in the residual resistivity at the metamagnetic transition. The multiple Fermi surfaces of $Sr_3Ru_2O_7$ might help to reduce the Coulomb energy of the charge distribution by mutual screening. Then the charge distribution of the passive bands or orbitals with a flat density of states near the Fermi level will tend to cancel the accumulated charge in the active magnetic bands. However, as the passive bands have a nonzero compressibility, a full compensation which would wipe out the inhomogeneity completely is unlikely.

In the liquid-gas transition the entropy jump is conserved by the Maxwell construction if the transition is crossed at a fixed pressure. In the metamagnetic case the total density is fixed during the transition. The Maxwell construction interpolates between the low magnetization below the transition and the high-m phase above. This transforms the step at h_m into a continuous rise between the lower and upper coexistence fields $h_<$ and $h_>$. Across the transition, a fraction $0<p<1$ will be already in the dense large-h phase corresponding to $n_>(h)$, while $1-p$ will remain in the low-h less dense phase with $n_<(h)$, and p increases from 0 to 1. Now, the Coulomb interaction will enforce charge neutrality on a nanoscale. It is natural to assume that the interface between the two phases and the remaining Coulomb energy due to the inhomogeneity costs some energy $g_I>0$. Taking this into account, the straight line for the energy of the phase-separated solution which undercuts the Gibbs potential of the homogeneous system between $n_<(h)$ and $n_>(h)$ gets shifted upward by g_I. Then nanoscale phase separation is energetically favorable only for p values between $p_{\min}$ and $p_{\max}$; otherwise, the homogeneous state will be lower in energy. When we compute the magnetization with this additional constraint $p_{\min}<p<p_{\max}$, we obtain two steps. One is near $h_<$, where p jumps from 0 to a nonzero $p_{\min}$. The other is near $h_>$, where the minority domains are squeezed out and p jumps from $p_{\max}$ to 1. Hysteresis is most likely at these two jumps; in between, the evolution is a continuous change of the relative size p of the two components and their densities. A precise calculation of g_I is difficult. In order to outline our idea, in Fig. 2(c) we assume a value $g_I=5\times10^{-7}W$ per unit area which is about 1/5 of the maximal energy gain of the inhomogeneous solution. Obviously, if g_I is too large, inhomogeneity does not occur.

In the experiments,[13] the resistivity anomaly becomes smaller when the in-plane component of the magnetic field is increased. Just as was suggested for the dFSD domains,[13] this could have the effect of orienting the density pattern. Then the transport along and orthogonal to the in-plane field should be different.

When the temperature is increased the magnetization jumps get smoothed out. As the spin symmetry is broken by $h\neq0$, there can be no phase transition associated with a symmetry breaking. However, the instability towards phase separation described above yields a thermodynamical distinction between the low-T micro-phase-separated state and the normal state at higher T. Above a threshold temperature T_{neg}, the negative curvature region in the Gibbs potential disappears. Hence the density remains homogeneous through the h range where m rises steeply. We speculate that T_{neg} is an upper bound for the temperature scale below which the T-dependent inelastic scattering becomes unobservable in the experiments. If the interface energy g_I is T independent, the inhomogeneity only sets in at $T<T_{\text{neg}}$. In Fig. 2(d) we show the inhomogeneity regions in the h-T diagram for $g_I=0$ and for a T-independent $g_I=5\times10^{-7}W$. Lifetime effects due to impurities act like increasing T and reduce the inhomogeneity region as well.

Experimentally, this scenario could be checked by local probes which are able to resolve spatial variations of the charge distribution—e.g., scanning tunneling microscopy. Note that in Ref. 14 the authors state carefully that phase separation cannot be ruled out near the transition.

IV. CONCLUSIONS

In conclusion, we analyzed two possible scenarios for the metamagnetic transition in the layered ruthenates. For the d-wave FS deformation picture,[13,14] a simple one-band t-t' Hubbard model can account for a charge instability that splits the saddle points of the dispersion only if the dominant magnetic fluctuations have a large wave vector connecting these saddle points. This does not seem to be the case in view of the many experimental and theoretical indications that strontium ruthenates are close to ferromagnetism. For dominant ferromagnetic fluctuations, we do not find an attractive coupling constant for the d-wave FS deformations.

As an alternative we extended the Stoner model of itinerant metamagnetism by Binz and Sigrist[8] and analyzed the possibility of microscopic phase separation. The metamagnetic transition in this model occurs in a region which is unstable with respect to density variations. Due to this general tendency, at first-order transitions the charge distribution might become inhomogeneous. We discussed the effects of the Coulomb repulsion and interface energies. If they do not cancel the energy gain by the phase separation completely, nanoscale inhomogeneity could account for the observed

anomaly in the residual resistivity and the occurrence of two steps in the magnetization.

Finally we note that Binz *et al.*[27] have considered a scenario with uncharged magnetic domains due to the demagnetization effect. We hope that further experiments can distinguish between the various proposals.

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[25] We can write $Un_\uparrow n_\downarrow = U[(n_\uparrow + n_\downarrow)^2 - (n_\uparrow - n_\downarrow)^2]/4$. The second term is the exchange term responsible for the magnetism. The first term penalizes charge inhomogeneity and is dropped in our treatment and in Ref. 8. We have checked that it reduces the phase separation tendency described later on only quantitatively.

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