Electronic phase separation and related inhomogeneity is ubiquitously seen in strongly-correlated systems. A typical example is found between ferromagnetic metal and antiferromagnetic insulator in CMR manganese oxides. Here we demonstrate that the geometrical frustration brings distinctive aspects into the phase separation phenomena. From Monte Carlo simulation and a simple energy comparison for the pyrochlore double-exchange model, we show that such phase separation takes place between ferromagnetic and paramagnetic metals. We discuss the relevance of our results to a spin-glassy metallic phase found in Mo pyrochlore oxides under external pressure.

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function of the distance in unit of the nearest-neighbor bond states. The data in (b)-(d) are for pyrochlore lattice. (c) and (d) Density of states for the two lengths. The data are measured along the 1D chains in the sizes $N_s = 16, 32, 64$, and 128 sites which correspond to $1 \times 1 \times 1$, $1 \times 1 \times 2$, $2 \times 2 \times 1$, and $2 \times 2 \times 2$ cubic unit cells of the pyrochlore lattice shown in the inset of Fig. 1(a). To suppress the finite-size effect, we take an average over the twisted boundary conditions which is a well-established technique to smear out the extrinsic “staircase-like” structure of the electron density as a function of the chemical potential. This averaging is efficient, in particular, to identify PS region, since it enables us to extract the intrinsic jump associated with PS even in small-size clusters, as demonstrated below.

Figure 1(a) shows a typical MC result for the electron density per site $n = \sum_i (\hat{c}_i^\dagger \hat{c}_i)/N_s$ as a function of the chemical potential $\mu$. It is clearly seen that the system-size dependence is suppressed even at the low temperature ($T$), owing to the averaging over the twisted boundary conditions. The results reveal two sharp jumps: One is from $n \approx 0.49$ to $n \approx 0.75$ at $\mu_{c1} \approx 1.4$, and the other is from $n \approx 0.04$ to $n \approx 0.16$ at $\mu_{c2} \approx -3.2$. These two jumps signal the electronic PS; the system is not stable at a density in these regions and phase-separated into two states at the edges of jump.

Both the PS take place between ferromagnetic and paramagnetic states. This is corroborated by the spin correlation plotted in Fig. 1(b). The results show typical behaviors on two sides of the PS at $\mu_{c1} \approx 1.4$. At $\mu < \mu_{c1}$, the spin correlation is large and converges to a positive value for farther neighbors, indicating a long-range ferromagnetic ordering. On the other hand, at $\mu > \mu_{c1}$, the spin correlation is AF for neighboring sites, but decays quickly to zero, suggesting a disordered paramagnetism.

The ferromagnetic and paramagnetic states are both metallic. Figures 1(c) and 1(d) present the density of states (DOS) per site for the two states. The results show that both states have no energy gap at the Fermi energy $\omega = 0$. This is consistent with the $\mu$ dependence of $n$ in Fig. 1(a): $\partial n/\partial \mu$ is nonzero on the both sides of PS, namely, the system is charge compressible. The ferromagnetic state is stabilized by the DE interaction, and hence, it is metallic. The paramagnetic state is also considered to remain metallic; a gap formation by a simple AF ordering is suppressed by strong frustration. All the above situations of the magnetic and electronic states hold in a similar manner for PS at a lower density at $\mu_{c2} \approx -3.2$, while in this case the system is the ferromagnetic (paramagnetic) state for $\mu > \mu_{c2}$ ($\mu < \mu_{c2}$).

We therefore conclude that the electronic PS occurs between the FM and PM states. This is a new type of PS appearing between metallic states, in sharp contrast with that in the unfrustrated case between FM and AFI with a clear energy gap. In the present model, we have shown in our previous paper a crossover from the simple PM to the cooperative PM (CPM) states; the crossover temperature generally goes to zero on the verge of PS because of the cancellation between DE ferro and SE AF interactions. Therefore, the electronic PS discussed here should in general occur above the crossover temperature and thus between FM and the simple PM.

The finite-$T$ phase diagram for PS is determined by keeping track of the discontinuity of $n(\mu)$. The results are shown for the higher-density PS at $\mu_{c1}$ for different values of $J_{AF}$ in Fig. 2. The PS region appears in a dome-like shape in the plane of $n$ and $T$, whose top connects to the second-order phase boundary between FM and PM. As increasing $J_{AF}$, the maximum temperature of PS is first enhanced slightly, but is suppressed for larger $J_{AF}$: PS disappears for $J_{AF} > 0.1$ in the $T$ range that we have...
calculated in MC ($T > 0.01$).

Now we try to understand the origin of PS on the basis of a simple argument on the energy comparison. Here we assume that FM is in the perfectly-ordered (fully-saturated) state and PM is in the completely-disordered state which is approximately obtained by using PM solution of the dynamical mean-field theory. In this assumption, we neglect not only the short-range AF correlations in the PM states, but also possibility of any other symmetry-broken states. We will see, however, that the energy comparison between the two simple states gives a reasonable estimate of PS regions in comparison with the finite-$T$ MC results.

Figure 3(a) shows the results for this analysis. The lowest curve in the figure is the energy for the FM state at $J_{AF} = 0$, which is essentially given by that for non-interacting spinless fermions. The FM state has two dispersive bands for $-6 \leq \omega \leq -2$ and $-2 \leq \omega \leq 2$, and two degenerate flat bands at $\omega = 2$. Because of the $\delta$-functional flat bands, the energy as a function of $n$ is given by a straight line from $n = 0.5$ to $n = 1.0$ in Fig. 3(a). DOS is shown in the inset with the one for the PM state. When we turn on $J_{AF}$, the perfectly-ordered FM state costs the SE AF energy, though the completely-disordered PM state does not because the SE energy is averaged to be zero. Figure 3(a) illustrates the situation for $J_{AF} = 0.04$ and 0.08. We can identify the PS region by drawing common tangent lines for the energy curves of FM and PM states (dashed lines in the figure). Note that PS appears in both small and large density regions for $J_{AF} > 0$.

We carry out the energy comparison with varying $J_{AF}$, and summarize the phase diagram shown in Fig. 3(b). In the middle of the phase diagram, we have the DE FM state, which shrinks as increasing $J_{AF}$ because of the loss of kinetic energy. The FM state is surrounded by the PS region between FM and PM. The PS has a finite width in all regions and a direct continuous transition from FM to PM is prohibited, except for a marginal point at $(n, J_{AF}) \simeq (0.3, 0.11)$.

Let us compare the energy argument and the finite-$T$ MC results. The phase boundaries obtained from the analyses in Fig. 3 are plotted as the filled circles at $T = 0$ in Fig. 2. The finite-$T$ MC phase boundaries appear to be extrapolated smoothly to the $T = 0$ results, except at $J_{AF} = 0$, where some discrepancy in the right boundary is seen. It is probably due to the singular $\delta$-functional DOS discussed in Fig. 3. The overall agreement supports that our simple energy comparison is not far from the reality and that the PS is essentially a metal-to-metal one from FM to PM.

This peculiar PS is not specific to the present model. We demonstrate it here by extending the model with including the next n.n. hopping $t'$, Figure 4 presents the results of the similar energy comparison for the extended model. For finite $t'$, the flat-band singularity in FM is removed, at the same time, the sharp cutoff at the band top in PM is rounded off [cf. the insets of Fig. 3(a)]. Nonetheless the metal-to-metal PS remains robust in a similar manner, as shown in the phase diagrams in Fig. 4. PS will be robustly observed even when the model is extended to describe more complicated band structure and orbital degeneracy.

We compare our results with those obtained in the absence of frustration. In the absence of frustration, PS takes place between FM and AFI as mentioned in the introduction. AFI appears at a commensurate filling, such as $n = 1.0$. The bandwidth of the AFI state is much smaller than that of FM, which drives the discontinuous change of the electron density associated with PS. Furthermore, the discontinuity is naturally expected since the magnetic transition between ferromagnet and antiferromagnet is necessarily of first order. In contrast to the unfrustrated case, our PS obtained for the frustrated pyrochlore model occurs between FM and PM. The se-
vere frustration suppresses the AF long-range order. In
the ground state, the DE-induced ferromagnetism bears
full saturation of moment to maximize the kinetic energy,
and hence, if there is a transition from FM to a param-
agnet by changing the chemical potential, the transition
becomes discontinuous with a jump of the moment. The
magnetic discontinuity naturally accompanies a discon-
inuous change of the electron density, which leads to
PS, as demonstrated in Figs. 9 and 11. The AF SE in-
teraction enlarges the paramagnetism to doped metallic
regions with reducing the FM region, resulting in the pe-
culiar metal-to-metal PS in our model. Hence the com-
plete polarization of the DE FM state at \( T = 0 \) plays a
key role in the present PS.

We note that the argument is in some sense precipitate
and the situation will be more complicated in the low-\( T \)
limit for large \( J_{AF} \) region, since the spin disordered state
might be unstable toward some symmetry breaking, such
as an incommensurate magnetic ordering. It is anticip-
pated in the present DE model that the kinetic motion of
electrons, which in general leads to farther-neighbor DE
interactions, acts as a degeneracy-lifting perturbation. In
fact, in the pyrochlore Heisenberg AF spin system, it is
known that any perturbation such as farther-neighbor
exchange interactions may force the system to order [19].

What we have revealed is that the \( T \) scale for such an-
ticipated phase transitions is very small in the present
DE model (at least, smaller than the \( T \) range reached
in the MC simulation) and the PS between FM and PM
takes place well above it. We expect that a similar PS
between, a spin-glassy metallic (SGM) state appears at low
\( T \). In the transition to SGM, there is a glassy response in
the AC susceptibility but no substantial anomaly in the
resistivity. This SGM is peculiar because a spin-glassy
behavior is observed in the insulating phase in the case of
CMR manganites. The spin-glassy insulating behav-
ior can be understood by competition between FM and
AFI with a formation of FM clusters in the matrix of
AFI (or charge/orbital ordered insulator). In the Mo py-
rochlores, the competing phases under pressure are FM
and PM, and therefore, we speculate that the interven-
ing SGM state originate from the electronic PS found in
our results. The glassy magnetic response is presumably
explained by domain-like structure or a mixed state; for
such situation, it is crucial to consider the effect of long-
ranged Coulomb interaction as well as quenched random-
ness [20, 21]. In the present case, the competing phases
are both metallic, and hence, we expect that the resulting
sgg3y state remains metallic. Further study by extend-
ing our model is necessary to confirm this scenario. It is
also interesting to extend the experimental study away
from the commensurate filling for comprehensive under-
standing of the phenomena.

Finally we comment on the relation between PS and
the flat band singularity at \( J_{AF} = 0 \). It was argued
that the DE FM is no longer stable when the chemi-
ical potential is in the flat band [22]. This instability
corresponds to the jump from FM at \( n = 0.5 \) to the
macroscopically-degenerate state at \( n = 1.0 \) in Fig. 3. A
jump of electron density associated with the flat band
singularity smoothly evolves to PS for \( J_{AF} > 0 \). This
suggests a possibility to understand the instability from
the viewpoint of the electronic PS. Further analysis is left
for future study.

To summarize, we have investigated the electronic
phase separation in the double-exchange model defined
on the frustrated pyrochlore lattice. The phase separa-
tion is found to occur between the ferromagnetic metal
and the paramagnetic metal: The former is stabilized by
the double-exchange interaction, and the latter is induced
by the frustrated antiferromagnetic super-exchange in-
teraction. We discuss that the phase separation ex-
plains the peculiar spin-glassy metallic behavior in Mo
pyrochlore oxides under external pressure. The metal-to-
metal phase separation is characteristic of the frustrated
double-exchange system, which is not seen in the unfrus-
trated models studied for the colossal magneto-resistive
manganites.

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Another possibility is the Anderson insulator, but it is unlikely in the clean system without randomness.

For very small $J_{AF}$, PS at low $n$ takes place between FM and the paramagnetic state with the absence of electrons, $n = 0$. Similar situation occurs in general, even at high $n$, as seen for the cases in the absence of the flat-band singularity in Fig. 4.