Dimensional trend in CePt$_2$In$_7$, Ce-115 compounds, and CeIn$_3$

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We present realistic Kondo-lattice simulation results for the recently-discovered heavy-fermion antiferromagnet CePt$_2$In$_7$ comparing with its three-dimensional counterpart CeIn$_3$ and the less two-dimensional ones, Ce-115’s. We find that the distance to the magnetic quantum critical point is the largest for CeIn$_3$ and the smallest for Ce-115’s, and CePt$_2$In$_7$ falls in between. We argue that the trend in quasi-two-dimensional materials stems from the frequency dependence of the hybridization between Cerium 4f-electrons and the conduction bands.

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Recently-discovered CePt$_2$In$_7$ [1, 2] has provided a new approach toward the two-dimensional (2D) limit in CeIn$_3$-derived heavy-fermion material family, among which Ce-115 materials have been discussed intensively in the past decade. In the hot debates on 115’s, the possible scaling of the superconducting transition temperature $T_c$ to $c/a$, where $a$ and $c$ are the lattice constants of the tetragonal crystal structure, has been discussed [3, 4] and there has been a hope to raise $T_c$ by making a more 2D-like material. In this context $c/a = 4.694$ (if $c$ should be taken to be the interlayer distance between Cerium planes, the scaling parameter should be taken to be 2.347) in CePt$_2$In$_7$ [2] looks promising as compared to the typical values of $c/a \sim 1.6$ in Ce-115’s where the highest $T_c$’s have been found among Cerium heavy-fermion compounds [3, 4].

Theoretically, the superconductivity (SC) with high $T_c$’s in strongly-correlated materials has been discussed to be mediated by magnetic fluctuations [8] and the possible mechanism for SC seems to be intimately related to the nearby magnetic quantum critical point (QCP) [8]. An empirical material-designing principle to get high $T_c$ would be to make it as close as to QCP and it is desirable for a given possibly high-$T_c$ material to know how close it is located to QCP. The robustness of the magnetic pairing has been discussed considering the spatial dimensional effects [10], stressing the stronger robustness in the more 2D-like systems even though care must be taken regarding the details of the electronic structure. Thus we are motivated to address QCP in CePt$_2$In$_7$ fully taking into account its electronic structure and see the trend among the related materials, namely, CeIn$_3$ with $c/a = 1$ and Ce-115’s with slightly larger $c/a \gtrsim 1.6$, to see if any microscopic origin of $c/a$-scaling of $T_c$ could be traced quantitatively to QCP. In the present work we predict the QCP’s for all of these materials to elucidate where exactly CePt$_2$In$_7$ is located in the neighborhood of QCP.

Recent experiments on CePt$_2$In$_7$ [11] have shown that this material is an ideal Kondo lattice with commensurate antiferromagnetism. It can be a good target for realistic Kondo lattice simulations [12] for which it has been shown that the description works fine in the Kondo limit, meaning that local $f$-level position, $\epsilon_f$, measured from the Fermi level should be negative and large, $U + \epsilon_f$ should be positive and large with $U$ being the on-site Coulomb repulsion energy, and at the same time hybridization $V^2$ should not be too big. When the valence fluctuations start to dominate the Kondo lattice simulations does not work and we have to go back to the lattice simulations based on the Anderson impurity problem [13], which would increase the computational cost. It has been discussed that Ce-115’s, especially CeRhIn$_5$ and CeIn$_3$, have well localized electrons [14] and we expect that the comparison between CePt$_2$In$_7$, CeRhIn$_5$/CeIrIn$_5$, and CeIn$_3$ in our simulations would make sense, while CeCoIn$_5$ might have to be looked at with some extra care.

We define our realistic Kondo lattice Hamiltonian starting with the first-principles electronic-structure cal-
solutions are numerically exact up to the approximation in the local QMC impurity solver [30–33]. We plug-in the realistic of-the-art continuous-time quantum Monte Carlo (CT-QMC) to dress the QCP in a semi-quantitative way utilizing state-electron basis [29] which enables us to reach low temperature splittings are set to be 0 eV for all of the materials. Relevant quantities in Kondo physics, taken from experiments referring to Ref. [25]. The structural parameters to be plugged into LDA are listed in Table I for the Cerium heavy fermion materials. The first two columns are LDA + Hubbard-I results for the local f-level position $\epsilon_f$ and the value of the hybridization function on the Fermi level. The last column shows the crystal-field splittings in the tetragonal structure.

| $\epsilon_f$ [eV] | $-\text{Tr}3\Delta(0)/\pi$ [eV] | $\Delta_1$ and $\Delta_2$ [meV] |
|------------------|---------------------------------|---------------------------------|
| 2D CePt$_2$In$_7$ | -1.81                           | 0.174                           | 8.6, 12.9$^a$ |
| CeCoIn$_5$       | -1.97                           | 0.205                           | 6.8, 25$^b$  |
| CeRhIn$_5$       | -1.90                           | 0.209                           | 5.9, 28.5$^c$|
| CeIrIn$_5$       | -1.95                           | 0.220                           | 5.26, 25.875$^c$|
| 3D CeIn$_5$      | -1.72                           | 0.239                           | 12$^d$  |

$^a$ = 100 [K] and 150[K], a rough estimate [20]. $^b$ Ref. [21, 22]. Another crystal-field scheme in Ref. [23] gives essentially the same results.

$^c$ Ref. [24]. The latest crystal-field schemes in Ref. [21] give the values close to these.

$^d$ Ref. [25]. This materials has the cubic structure which brings $\Delta_1 = \Delta_2$.

Now we describe how we solve the realistic Kondo lattice model (KLM) that we have defined from $-3\Delta(\omega)/\pi$ in Fig. 1 and Table I. We use dynamical mean-field theory (DMFT) [27, 28] that is formulated on a local f-electron basis [28] which enables us to reach low temperature region with a modest computational cost and address the QCP in a semi-quantitative way utilizing state-of-the-art continuous-time quantum Monte Carlo (CT-QMC) impurity solver [30, 33]. We plug-in the realistic crystal-field and spin-orbit splittings as given in Table I in the local 4f-level in the impurity problem. Thus our solutions are numerically exact up to the approximation of DMFT. We look at the magnetic phase transition as a function of temperature and restore the Doniach phase diagram [34] by varying the Kondo coupling $J_K$ for a given material. Thus a magnetic QCP is found on a realistic Doniach phase diagram spanned by the Kondo coupling and temperature, and the realistic data point for the given material is picked up for the realistic value of $U = 5$ [eV] and $J_{\text{Hund}} = 1$ [eV] to estimate its distance to QCP.

Now we show how we determine the QCP of CePt$_2$In$_7$. The calculated temperature dependence of the inverse of the staggered magnetic susceptibility, $1/\chi(\pi)$, for CePt$_2$In$_7$ is shown in Fig. 2. Here we have employed random dispersion approximation [32] to estimate the two-particle Green’s function by decoupling, which would enhance our transition temperatures on top of DMFT as will be seen below. Being consistent with Doniach’s picture [34] where the winner of the competition between the magnetic-ordering energy scale $\propto J_K^2\rho$ and the Kondo-screening energy scale $\propto \exp[-1/(J_K \rho)]$ interchanges at some finite $J_K$, it is seen that small $J_K$’s give a diverging $\chi(\pi)$ at a finite Néel temperature $T_N$ while large $J_K$’s give a saturating $\chi(\pi)$ at low temperatures, and some value of $J_K$ in between gives the quantum critical point where $T_N$ vanishes. The Néel temperatures for smaller $J_K$’s are determined by linear extrapolation of $1/\chi(\pi)$ to the lowest temperature region and thus obtained $T_N$ is plotted against the Kondo coupling in Fig. 3 in a format of restored Doniach phase diagram. Here we note that we just identified what we call QCP by a parameter segment where the finite Néel temperature seems to have vanished, and there is always a possibility that in some smaller parameter segments there is actually a coexistence region. There is also a possibility numerically for a
first-order phase transition where the Néel temperature actually jumps from a finite value to zero at a certain point of $J_K$. We leave the exact characterization of what we also call QCP here for future investigations and for the moment we would be satisfied with that it looks like QCP practically in most cases with a very small jump numerically, if any.

For CePt$_2$In$_7$, the realistic data point is at $J_K = 1.165 J_{K,0}$, which corresponds to $U = 5$ [eV] and $J_{\text{Hund}} = 1$ [eV], gives the Néel temperature $T_N \approx 10$ [K] which is larger than the experimental result $T_N = 4.5$ [K] due to our mean-field argument with DMFT and the random dispersion approximation which becomes exact on a lattice with perfect nesting. The same analyses are applied to the other materials to give the Néel temperature $T_N = 19$ [K] for CeIn$_3$, 15 [K] for CeRhIn$_5$, and 14 [K] for CeCoIn$_5$. Again the experimental values ($T_N = 10.2$ [K] for CeIn$_3$, 3.8 [K] for CeRhIn$_5$\cite{32,33} comes below these results. The data for CeCoIn$_5$ is not consistent with the experimental fact that CeCoIn$_5$ is a non-magnetic heavy fermion material\cite{32}. However the arrangement of the materials around QCP reveals that the apparent finite $T_N$ actually comes from being in an immediate proximity to QCP as shown in Fig. 4. We use a rescaled Kondo coupling on the horizontal axis in terms of its value right on the QCP, $t \equiv (J_K - J_{K,QCP})/J_{K,QCP}$, to remove a problem with the Doniach phase diagram that each material has its own energy scales.

Now we inspect the distance to the QCP of CePt$_2$In$_7$ referring to those of Ce-115’s and CeIn$_3$. The cubic parent material CeIn$_3$ is seen to be most separated from QCP and Ce-115’s are found to be concentrated in the neighborhood to QCP, with CeRhIn$_5$ on the magnetic side and CeIrIn$_5$ on the non-magnetic side. Our numerical resolution is not sufficient to locate CeCoIn$_5$ in its correct non-magnetic side, but it is clear that it sufficiently works to estimate the extreme closeness to QCP of Ce-115’s. We note that our calculation scheme might not be as good for CeCoIn$_5$ as for the others due to the possibly stronger effects of valence fluctuations in this material. It is seen that when we try to reach the quantum critical point (QCP) from CeIn$_3$ in the three-dimensional limit, CePt$_2$In$_7$ is located in the midway toward Ce-115’s which are located closest to the QCP. Seen from QCP, CeIn$_3$ is already close enough to enable the pressure-driven superconductivity\cite{34} and CePt$_2$In$_7$ would also have one. Making a material more 2D indeed helps it to come closer to QCP, which is reasonable in the general context that the lower spatial dimensionality would suppress the magnetic long-range order. However within the 2D-side, the trend among CePt$_2$In$_7$ and Ce-115’s is somewhat non-monotonic. The possible reason is seen in Fig. 4 where a dip around the Fermi level is seen for CePt$_2$In$_7$ which would help to drive it to the magnetic side with the re-
duced Kondo screening. On top of the spatial dimensionality, the frequency dependence of the hybridization seems to introduce the nontrivial trend in this way.

Here we note that precisely speaking we are discussing in the infinite-dimensional limit with the DMFT, but at least a semi-quantitative trend of $T_c$ would be satisfactorily addressed. We also note that in the literature the importance of momentum dependence of the hybridization has been stressed [18, 37, 38] which we have neglected. What we have seen is that the energy dependence seems to be sufficient at least to capture the trend in the distance to QCP, thus for the prediction of magnetically-mediated superconductivity with a possible high $T_c$.

In order to reach QCP, it would be interesting to have a material analogous to CePt$_2$In$_7$ without a big dip in the hybridization around the Fermi level. For that, the following possible ways for the material designing could help: 1) electronic carrier doping, 2) ascending T in the periodic table for CeT$_2$In$_7$ to enhance the hybridization, and 3) shifting T to the left- or the right-hand side on the periodic table to lift the dip off the Fermi level to enable the stronger Kondo coupling to drive the material toward the QCP side.

Finally we discuss the degree of the delocalization of $f$-electrons by looking at their contribution to the Fermi surface of the conduction electrons in our realistic Kondo lattice description for each material. Even if we have only localized $f$-electrons in our Hamiltonian, they contribute to the formation of the large Fermi surface via the hybridization and in this sense they can be delocalized [39]. We follow the procedure used in Ref. [40]. From our simulations for a give material at a fixed temperature, we get the conduction-electron self-energy $\Sigma(i\omega_n)$ and then track the temperature dependence of the real part of it at $i\omega_n = 0$ to look at the shift of the Fermi level. Here $\omega_n = (2n+1)nT$ is the fermion Matsubara frequency. The result is plotted in Fig. 5 It is seen that CePt$_2$In$_7$ has the most localized $f$-electrons down to the temperature range of 20 [K] among these materials while the 115’s have the strongest delocalization. CeIn$_3$ falls in between. We note that the trend in the $f$-electron delocalization does not exactly follow that in the hybridization strength right on the Fermi level seen in Fig. 1 and it is the outcome of the frequency dependence of the hybridization that we have taken into account in our realistic Kondo lattice simulations.

To conclude, we have predicted the QCP of CePt$_2$In$_7$ and discussed the possible strategy to have the higher $T_c$ in the related material within our realistic Kondo lattice description which has been shown to predict the properties of Ce-115’s and their parent material CeIn$_3$ semi-quantitatively. Our method’s predictability of magnetic QCP would further provide the material designing principle toward more high-$T_c$ materials in the upcoming material exploration.

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