Tunable magnetism of hexagonal Anderson droplet on the triangular lattice

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Motivated by recent progress on quantum engineered Kondo lattices, we numerically investigate the local magnetic properties of a hexagonal Anderson impurity droplet consisting of multiple rings of magnetic atoms periodically arrayed on a triangular lattice. We demonstrated the tunability of the magnetic properties via their evolution with the droplet geometry for two types of systems with distinct local orbital occupancy profile. We found that the local susceptibility of the central impurity of some types of droplets can be remarkably enhanced in contrast to the conventionally rapid decrease due to spin correlations of surrounding impurity rings. This unusual behavior is attributed to the charge fluctuation and its interplay with spin degrees of freedom. Our simulations complements the exploration of the novel artificial tunability of engineered lattice systems.

There has been a recent paradigm shift in the investigation of strongly correlated electronic systems from real materials to the atomic-scale manipulation of artificial lattices and/or superlattices [1][8]. For example, in the context of the Kondo physics in heavy fermion materials [9], the realization of artificial lattices has provided a radically new platform to both explore and manipulate the emergence of strong correlation effects. The resulting many-body phenomena at the nanoscale permit the diverse opportunities for studying the interplay between different degrees of freedom in a controllable manner. In particular, the quantum engineering of nanoscopic Kondo droplets has been demonstrated to be capable to coherently control the droplet’s properties such as its Kondo temperature [1]. Theoretically, they employed large-N expansion for the treatment of triangular Cu (111) surface lattice, which allows for two types of hexagonal magnetic atom droplets. Recently, the requisite conditions of the coherent Kondo lattice behavior for periodically arranged magnetic moments on a square lattice within the particle-hole symmetric Kondo lattice model (KLM) has been investigated [10].

Here we numerically explore the local magnetic properties of a hexagonal Anderson droplet consisting of multiple rings of impurity atoms on a triangular lattice in the framework of the Anderson lattice model, which has richer physics than the Kondo-lattice model because of the intrinsic charge fluctuations of f-electrons. Our focus is the evolution of the local properties within the droplet and more importantly their dependence on the spatial structure of the droplet.

We employ the two-dimensional Anderson droplet model (ADM) in the half-filled form on a triangular lattice

$$\mathcal{H} = -t \sum_{ij,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + c_{j\sigma}^\dagger c_{i\sigma}) - V \sum_{i \in D,\sigma} (c_{i\sigma}^\dagger f_{i\sigma} + f_{i\sigma}^\dagger c_{i\sigma})$$

$$+ U \sum_{i \in D} (n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2}) - \mu \sum_{i\sigma} n_{i\sigma} - \sum_{i \in D,\sigma} n_{i\sigma}$$

where $c_{i\sigma}^\dagger (c_{i\sigma})$ and $f_{i\sigma}^\dagger (f_{i\sigma})$ are creation(annihilation) operators for the conduction and local f-electrons on site $i$ with spin $\sigma$, respectively. $n_{i\sigma}^c,f$ are the associated number operators. $t = 1$ set to be the energy unit is the hopping amplitude between conduction electrons on nearest-neighbor sites $(ij)$ of a triangular lattice. $U$ denotes the local repulsive interaction for f-electrons and $V$ is the hybridization between the conduction and f-electrons. The chemical potential $\mu$ controls the average density of the system. The set $D$ defines the set of sites where the impurity droplet resides. Figure 1 illustrates the geometrical structure of our hexagonal lattice with open boundary and two types of droplets, where the distance between the consecutive impurity rings can be $n a_0$ and b) $\sqrt{3} n a_0$ separately with $a_0 \equiv 1$ the lattice constant and $n$ is integer. This geometry has direct relevance to recent study on the artificially engineered Kondo droplet system [11]. From now on, we denote the spatial structure of the droplet by $A/B\{n : N_r\}$ with $n$ the distance between consecutive rings and $N_r$ the number of rings such that Fig. 1 shows $A\{1 : 5\}$ and $B\{1 : 4\}$ droplets. With this notation, for example, the first ring of $A\{3 : 2\}$ droplet has the same location as the third ring of $A\{1 : 4\}$ droplet.

Although the ADM model breaks the translational symmetry of the lattice, it has close relation to the well-known periodic Anderson model (PAM), which is
conventionally believed to capture the essential physics of heavy-fermion materials [19]. Accordingly, PAM has been extensively explored numerically in various contexts, for example, of the phase diagram [11][12], universal Knight shift anomaly [13], d-wave superconductivity [14], Mott metal-insulator transition [15][17] etc. ADM can also be viewed as a special form of depleted PAM [18] and has relevance to the PAM with impurities [19][20].

The phase diagram of the PAM on triangular lattice has been explored extensively in the past decades, which hosts richer phases than its counterpart on square lattice [21][22]. Following the phase diagram reported in [24], we focus on the characteristic intermediate coupling strength \( U/t \approx 1.0 \) and \( c-f \) hybridization strength \( V/t \approx 1.0 \) such that they are of the same order of magnitude. To treat these energy scales on the equal footing, we solve the ADM by means of the conventional finite temperature determinant Quantum Monte Carlo (DQMC) [23]. Note that our hexagonal shape triangular lattice with open boundary is highly inhomogeneous and non-periodic so that both the conduction and \( f \) electron density distributions will affect the local properties throughout the lattice. In this work, we have treated with two types of systems: (1) the whole system is half-filled \( \rho = 1 \) by tuning the chemical potential \( \mu \) and (2) the droplet impurities are almost half-filled by setting fixed \( \mu = 0 \) for all droplet geometries. We emphasize that these two cases, which is equivalent in the conventional PAM on square lattice, differ in our lattice geometry. In the former system, both the conduction and \( f \) electron densities are away from and can exceed half-filling. In contrast, the latter system partially removes the charge fluctuations of \( f \)-electron whose density is almost half-filled while the conduction electron below half-filling presents mostly the spatial density modulation. Besides, to study a large enough lattice at low enough temperature with manageable sign problem and computational cost, most of results presented are for lattices with boundary length \( L = 10 \) such that the total number of sites is \( 3L^2 + 3L + 1 = 331 \), which has constrains on the maximal possible number of impurity rings for a particular droplet ring distance \( n \).

The key quantity is the local magnetic susceptibility defined as

\[
\chi_{ab}(r) = \int_0^\beta d\tau [n^a_{\tau}(r) - n^a_{\tau=0}(r)][n^b_{\tau}(0) - n^b_{\tau=0}(0)]
\]

with \( a,b \) denoting the conduction and \( f \) electron respectively and \( r \) is the location of the characteristic impurity along the red examination line in Fig. 1 on the \( r \)-th impurity ring so that the central impurity is at \( r = 0 \). Note that this red examination line passes the lattice corner and the middle of the lattice boundary for \( A \) and \( B \) type of droplets separately. We mention that the reduced number of nearest impurities so that differing spin correlation via RKKY interaction and also different neighboring conduction electron sea so that the Kondo screening will largely affect the the properties near the lattice boundary [10]. In some cases the following results will cover the data near or at the boundary despite that is not our focus apparently.

Figure 3 illustrates the evolution of the local \( f \)-electron susceptibility \( \chi_{ff}(r) \) versus \( r \) for various \( A \) and \( B \) droplet geometries in both systems of \( \rho = 1 \) and \( \mu = 0 \), where we have simplified the label as \( A/B(n : N_r) \) for various cases of \( N_r \). To access the large enough droplets (especially for \( A1 \) droplet), the temperature is chosen to be \( T = t/10 \), which is low enough to identify the essential properties presented here. The major features persist at a lower temperature \( T = t/20 \) with a smaller largest droplet size (\( N_r = 5 \) is the largest accessible \( A1 \) droplet due to the sign problem).

Firstly, \( \chi_{ff}(r) \) in both systems of \( \rho = 1 \) and \( \mu = 0 \) show oscillating behavior (the most clearly for \( A1 \) droplet) with increasing \( r \) for \( A/B(n : N_r > 2) \) droplets before reaching the outermost ring, where this `regular' oscillation breaks down due to the lattice boundary effect mentioned before or a similar effect occurred at the droplet boundary [10]. Neglecting the droplets, e.g. \( A1:9 \), whose outermost ring approaches to or locates at the lattice boundary, we note that \( A1 \)'s outermost ring has an upturn of \( \chi_{ff} \) while other droplets, e.g. \( A2:3 \) (\( \rho = 1 \) system) and \( A3:2 \) (\( \mu = 0 \) system), can have the opposite trends. This complication at the outermost ring stems from the density fluctuation (dominantly at the spatial region of the droplet boundary) of the system, which intertwines with the lattice and/or droplet boundary effects. In fact, the local density fluctuation (not shown), which mostly comes from the conduction electrons especially in \( \mu = 0 \) systems, and the oscillations of \( \chi_{ff}(r) \) are almost anti-correlated.

Secondly, the oscillation of \( \chi_{ff}(r) \) gradually diminishes with increasing \( N_r \) in the droplet size, as most clearly shown for \( A1 \) droplet of \( \rho = 1 \) system. In other words, the droplet’s central region becomes more and more homogeneous and coherent [24]. For other droplets with larger distance \( n \) between consecutive rings, we are limited by the lattice size to identify the diminishment of \( \chi_{ff}(r) \).

Thirdly, the weaker dependence of \( \chi_{ff}(r) \) on \( N_r \) for \( B \)-type droplets compared with their \( A \)-type counterparts signifies the impact of the differing geometrical arrangement of impurities, in particular, the distance between consecutive rings and the absence of nearest-neighbor impurities in \( B \)-droplets. In the sense of the minimal hopping distance between the consecutive rings, \( B1(\text{or} B2) \) droplet is more similar to \( A2(\text{or} A4) \) droplet.

The major difference between systems of \( \rho = 1 \) and \( \mu = 0 \) lies that \( \chi_{ff}(r) \) in the latter system gradually saturates with \( N_r \) while in the former system it remains growing even for large \( N_r \). This originates from the stronger charge effect in the half-filled system. Taking
A1 droplet for example, the site-dependent density keeps decreasing and approaches to half-filling upon increasing $N_r$ so that $\chi_{ff}(r)$ keeps growing in the former system; while the density saturates for large enough $N_r$ droplet in $\mu = 0$ systems. The stronger charge effects have to be suppressed to some extent by lowering the temperature, which has been verified at $T = t/20$ despite of the limitation of accessing only smaller droplet size. We will further discuss this issue later. At this point, we emphasize that the ‘full’ suppression of charge effects requires pushing to much lower temperature, which is difficult, if not possible, in our DQMC simulations. Kondo lattice model is more appropriate in this regard. In summary, the rich behavior of $\chi_{ff}(r)$ dependent on the droplet geometries indicates the possibility of artificial manipulation of the magnetic properties in a controllable manner.

One fascinating feature of $\chi_{ff}(r)$ is the strong dependence of $\chi_{ff}(r = 0)$ at the central impurity on the droplet geometry. Figure 3 illustrates its evolution upon increasing $N_r$ for various types of droplets. $\chi_{ff}(r = 0)$ reflects the competition between inter-impurity antiferromagnetic spin correlation with the surrounding impurity rings mediated via RKKY interaction and Kondo screening of the conduction electrons. Moreover, the charge fluctuations in our systems interplay with these two factors to complicate the whole picture. Fig. 3(a-b) compare the behavior of $\chi_{ff}(r = 0)$ between two types of systems at $T = t/10$ and (c-d) present the comparison at lower temperature $T = t/20$. For $A$-type droplet with smallest distance between rings e.g. $n = 1$, $\chi_{ff}(r = 0)$ decreases rapidly from its single-impurity value and keeps growing in (a) while gradually saturates after an oscillating behavior in (b) with increasing $N_r$. As we discussed previously, this difference stems from the stronger charge effects in $\rho = 1$ systems, which can be partially suppressed at lower temperature $T = t/20$ shown in (c). This saturation similar to that reported for the KLM on square lattice [10] can be attributed to the buildup of spin correlations induced from the neighboring droplet rings, which gives rise to the collective-like screening of

FIG. 2: Local f-electron susceptibility $\chi_{ff}(r)$ for various A and B droplet geometries in both systems of $\rho = 1$ and $\mu = 0$, where we have simplified the label as $A/Bn \equiv A/B\{n : N_r\}$ for various cases of $N_r$. $T = t/10$ is chosen to access large enough droplets (especially for A1 droplets).

FIG. 3: Comparison between $\chi_{ff}(r = 0)$ at the central impurity as a function of the number of impurity rings $N_r$ for various A and B droplets.
the central impurity. As pointed out in Fig. 2, $\chi_{ff}(r = 0)$ in $B$-type droplets has more moderate dependence on $N_r$. The moderate deviation from the single impurity case implies the compensation between local Kondo, inter-impurity spin correlation, and charge fluctuation effects. Unfortunately, it is impossible to access much larger lattice and/or lower temperature to identify the saturation of all droplet geometries, especially the $B$-type with larger distance between rings.

The most unusual feature due to the interplay between various intertwined effects manifests when increasing the distance $n$ between impurity rings, where the fate of the decrease of $\chi_{ff}(r = 0)$ upon $N_r$ is significantly modified. In fact, $\chi_{ff}(r = 0)$ can even be enhanced in $A_3, A_4, B_3$ droplets in $\rho = 1$ and $A_2, B_1$ droplets in $\mu = 0$ systems (a-b), which persists at lower temperature $T = t/20$ (c-d). As verified via the density profile, this remarkable enhancement implies the significant charge redistribution, which in turn results in the tunable magnetic susceptibility for a particular impurity embedded in a system.

To further understand the nontrivial dependence of the local magnetism upon the droplet geometry, we also explored $\chi_{ff}(r = 0)$ for droplets with a single impurity ring $N_r = 1$ but varied distance $n$ to the central impurity. As can be seen in Figure 4(a-b), it oscillates periodically with $n$ in both $\rho = 1$ and $\mu = 0$ systems although the lattice size forbids accessing more oscillations for $B$-type droplets, which vividly depicts the tunability of the magnetic properties via modulation of the droplet geometry. The strong charge effect is illustrated in Fig. 4(c-d), which displays the anticorrelation between the local density profile at the lattice center and $\chi_{ff}(r = 0)$. Although the charge fluctuation for $f$-electron can be partially suppressed by enforcing its nearly half-filled occupancy via $\mu = 0$, the density profile of local conduction electrons plays an important role in determining the magnetic properties of the droplet impurities.

Finally, we remark on the relevance of our results to the latest exploration of Co adatoms on Cu(111) surface [1]. It is not straightforward to compare to this earlier report employing large-$N$ expansion technique based on KLM due to (I) the intrinsic difficulty of extracting the local hybridization strength via the analytical continuation of local interorbital Green’s function; (II) the strong charge effects in our ADM; and (III) the finite size effect in contrast to the essentially infinitely large Cu(111) surface [1]. Further design of the lattice and/or droplet setup are requisite to perform insightful direct comparison. We mention that the interorbital magnetic susceptibility $|\chi_{ff}(r)|$ is found to largely anticorrelates with $\chi_{ff}(r)$ presented in Fig. 2 which naturally reflects the local competition between the Kondo screening and inter-impurity spin correlation.

In conclusion, we have employed the numerically exact DQMC simulations in the framework of Anderson droplet model to investigate the local magnetic properties associated with the hexagonal impurity droplet embedded in a triangular lattice. We demonstrated the tunability of the magnetic properties by the evolution of local susceptibility with the droplet geometry for two types of systems with differing local orbital occupancy profile. Our ADM has the intrinsic intertwined spin and charge degrees of freedom, whose fluctuations largely affect the local magnetic properties, although the charge fluctuation for $f$-electron can be partially suppressed by enforcing its nearly half-filled occupancy. The natural extension of the present work is to investigate the Kondo droplet model [10] with the absence of $f$-electron charge fluctuation, and/or with more appropriate lattice band fillings setup. Besides, the Kondo holes in a droplet deserves further exploration [11, 25, 20]. Despite that the direct comparison with the recent progress on quantum engineered Kondo lattice is not straightforward, our simulations complements the exploration of the novel artificial tunability of engineered lattice systems.

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