Kondo Quantum Criticality of Magnetic Adatoms in Graphene

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We examine the exchange Hamiltonian for magnetic adatoms in graphene with localized inner shell states. On symmetry grounds, we predict the existence of a class of orbitals that lead to a distinct class of quantum critical points in graphene, where the Kondo temperature scales as $T_K \propto |J - J_c|^{1/3}$ near the critical coupling $J_c$, and the local spin is effectively screened by a super-ohmic bath. For this class, the KKKY interaction decays spatially with a fast power law $\sim 1/R^2$. Away from half filling, we show that the exchange coupling in graphene can be controlled across the quantum critical region by gating. We propose that the vicinity of the Kondo quantum critical point can be directly accessed with scanning tunneling probes and gating.

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Graphene is a single atomic sheet of carbon atoms with elementary electronic quasiparticles that behave as massless Dirac fermions\cite{1}. The Kondo effect has been recently observed in graphene\cite{2, 3} and the formation of a Kondo screening cloud around a magnetic adatom is quantum critical at half filling\cite{1, 4}, crossing over at weak coupling to the standard Fermi liquid case, when the DOS is locally restored by disorder\cite{1} or gating effects\cite{5}. The Kondo resonance in graphene is also strongly sensitive to the position of the adatom in the honeycomb lattice, where the interplay of orbital and spin degrees of freedom may give rise to an SU(4) Kondo effect\cite{6}.

In this letter, after establishing a generic one-level exchange interaction Hamiltonian for magnetic adatoms in graphene, we show there is a symmetry class of orbitals that lead to a fixed point where the Kondo effect is described by the single impurity Anderson Hamiltonian\cite{7, 8, 9},

$$
\hat{H}_g = \hat{H}_d + \hat{H}_f + \hat{H}_U + \hat{H}_V,
$$

where $\hat{H}_U = \sum_{\sigma} \epsilon_0 \hat{n}_{\sigma} \sigma$ is the Hamiltonian of the localized electrons, with $\hat{n}_{\sigma} = \hat{n}_{\sigma} \sigma$ as the number operator, and $\epsilon_0$ is the energy of the local state measured relative to the Dirac point, $\hat{U} = U \hat{n}_{\sigma} \sigma$ gives the electronic repulsion in the localized level, and $\hat{H}_V$ describes the hybridization between the local level and the graphene electrons.

The adatoms in graphene can sit for instance on top of a carbon atom, where the hybridization Hamiltonian is $\hat{H}_V = V \sum_{\sigma} \hat{n}_{\sigma} \sigma$, or in the hollow site in the center of the honeycomb hexagon, where the adatom hybridizes with the two sublattices, $\hat{H}_V = \sum_{\sigma} \sum_{i=1}^3 [V_{a,i} \hat{n}_{\sigma}(\mathbf{a}_i) + V_{b,i} \hat{n}_{\sigma}(-\mathbf{a}_i)] \sigma$, with $V_{x,i}$ as the hybridization strength of the localized orbital with each of the three surrounding carbon atoms sitting on a given sublattice. In momentum representation\cite{9},

$$
\hat{H}_V = \sum_{\mathbf{p},\sigma} \left[ V_{a,\mathbf{p}} \hat{b}_{\sigma,\mathbf{p}}^\dagger + V_{b,\mathbf{p}} \hat{b}_{\sigma,\mathbf{p}} \right] \sigma + h.c.,
$$

where $V_{a,\mathbf{p}} \equiv V$, and $V_{b,\mathbf{p}} = 0$ for a top carbon site, say on sublattice $A$ ($A$-site). When the adatom sits in

![Figure 1: Representation of (a) an s-wave and (b) an f-wave orbital, when the adatom sits in the center of the graphene honeycomb hexagon. (c) Substitutional impurity in a single vacancy. In the three cases, the adatom hybridizes equally with the neighboring carbons on the same sublattice.](image)
the center of the hexagon (H-site), or else for a substi-
tutional impurity in a single vacancy (S-site), the
hybridization function is $V_{s,p} = \sum_{i=1}^{3} V_{s,i}e^{ip\cdot a_i}$. On H-
sites, for an s-wave orbital, $V_{s,i} = V$, giving $V_{s,p} = V\phi_p$, whereas for an in-plane f-wave orbital, as shown in
Fig. 1b, where the orbital is odd in the two sublattices, $V_{s,i} = -V_{s,i} = V$, resulting in $-V_{s,p} = V_{s,p} = V\phi_p$. In
the case of an s or in-plane f-wave orbital on an S-site
on sublattice $A$, $V_{s,p} = 0$, and $V_{s,p} = V\phi_p$, whereas for a substitutional impurity on a B-site, $V_{s,p} = V\phi_p$, and $V_{s,p} = 0$ (see Fig. 1c).

Diagonalizing the non-interacting part of the Hamil-
tonian $H$ in the $A$, $B$ sublattices,

$$H_g = \sum_{p,\sigma} E_\alpha(p)c_{\alpha,\sigma,p}^\dagger c_{\alpha,\sigma,p}, \quad (3)$$

where $E_\alpha(p) = \alpha|\phi_p|$ is the graphene tight-binding spectrum, with $\alpha = \pm$ labeling the conduction and valence bands, and $c_{\pm,\sigma,k} = ((b_{\nu,k} \pm \phi_{\nu}^\dagger)/|\phi_k|a_{\nu,k})/\sqrt{2}$ are the new operators in the diagonal basis. The hybridization term in the rotated basis is

$$H_V = V \sum_{\alpha=\pm} \sum_{p,\sigma} [\Theta_{\alpha,p} c_{\alpha,\sigma,p}^\dagger f_\sigma + h.c.], \quad (4)$$

where

$$\Theta_{\alpha,p} = (V_{b,p} + \alpha V_{s,p}^* \phi_{p}^\dagger |\phi_p|)/(\sqrt{2}V). \quad (5)$$

In particular, $\Theta_{\alpha,p}^A = 1/\sqrt{2}$ when the adatom is on top of an A-site, $\Theta_{\alpha,p}^B = \alpha \phi_{p}^\dagger |\phi_p|/(\sqrt{2})\phi_p$ on a B-site, and $\Theta_{\alpha,p}^{H,\gamma} = |\phi_p| + (-1)^{\gamma} \alpha \phi_{p}^\dagger |\phi_p|/(\sqrt{2})\phi_p$ when the adatom sits on an H-site, where $\gamma = 0$ for an s-wave orbital and $\gamma = 1$ in-plane f-wave orbital. In the substitutional case, $\Theta_{\alpha,p} = \phi_{p}/\sqrt{2}$ for an impurity on sublattice $A$, and $\Theta_{\alpha,p}^{B} = \alpha \phi_{p}^\dagger/(\sqrt{2})\phi_p$ on sublattice $B$.

For all possible symmetries, the orbitals of adatoms
sitting on S or H sites can be classified among those that either break or preserve the $C_{3v}$ point group symmetry of the triangular sublattice in graphene. Since $|\phi_p|$ scales with $|\omega|/t$, the orbital level broadening, $\Delta(\omega) = \pi V^2 \sum_{\alpha,p} \Theta_{\alpha,p}^2 \delta(\omega - \alpha|\phi_p|)$ is either $\Delta(\omega) \propto V^2 \rho(\omega)$ for orbitals that explicitly break the $C_3$ point group symmetry, in which case $\Theta_{\alpha,p}$ scales to a constant near the Dirac points, where $\rho \propto |\omega|$ is the graphene density of states (DOS), or else $\Delta(\omega) \propto V^2 \rho(\omega)|\omega|^2/t^2$, for $C_{3v}$ invariant orbitals, when $|\Theta_{\alpha,p}| \propto |\phi_p|$ scales to zero at small energy. The first class of orbitals, where $\Delta(\omega) \propto |\omega|$ (say, type I), represents the standard case of ohmic dissipation, and is described for instance by adatoms on top carbon sites, by $E_1(d_{xz},d_{yz})$ and $E_2(d_{xy},d_{x^2-y^2})$ representations of d-wave orbitals and $f_{xz}, f_{yz}, f_{x^2-y^2}$ orbitals in H/S sites. The second class, where $\Delta(\omega) \propto |\omega|^3/t^2$ (type II), represents a new class of super-ohmic dissipation, and is described by $s, d_{zz}, f_{z^2}, f_{x(x^2-3y^2)}$, and $f_{y(3z^2-y^2)}$ orbitals in H or S sites (see Fig. 1), where the adatom hybridizes equally with the three nearest carbon atoms on a given sublattice. On physical grounds, this new class emerges from quantum mechanical interference between the different hybridization paths in the honeycomb lattice, as the electrons hop in and out of the localized level. As we will show, these two classes of orbitals are described by two distinct types of Kondo QCP.

The Anderson Hamiltonian in graphene can be sepa-
rated in two terms, $H = H_0 + H_V$, and then mapped into a spin exchange Hamiltonian through a standard canonical transformation, $\hat{H} = e^{S}\hat{H}e^{-S} = \hat{H} + [S,\hat{H}] + \frac{1}{2}[S,[S,\hat{H}]] + \ldots$, where $S = -\sum_{p,\alpha,\sigma} V[1 - \hat{n}_{\alpha,\sigma} - \alpha|\phi_p|]^{-1} + \hat{n}_{f,-\sigma}(\epsilon_0 - \alpha|\phi_p|)]^{-1} \Theta_{\alpha,p} c_{\alpha,\sigma,p} f_\sigma - h.c.$, which results in a Hamiltonian that is quadratic in $V$ to leading order, $H = H_0 + [S,\hat{H}V]/2 + V(\phi_{p}^\dagger)(\phi_{p})$. At large $U$, the exchange Hamiltonian is given by

$$H_e = -J \sum_{kk'\alpha\alpha'} \Theta_{\alpha,k}^* \Theta_{\alpha',k'} S \cdot c_{\alpha,k}^\dagger c_{\alpha',k'} \sigma c_{\alpha,k}, \quad (6)$$

where $\sigma = (\sigma_1, \sigma_2, \sigma_3)$ are Pauli matrices and

$$J(\mu) \approx V^2 U^2/[(\epsilon_0 - \mu)(\epsilon_0 + U - \mu)] < 0, \quad (7)$$

is the exchange coupling defined at the Fermi level, $\mu$.

The validity of the exchange Hamiltonian is controlled by the ratio $\Delta(\epsilon_0)/|\epsilon_0 - \mu| < 1$, when the valence of the localized level is unitary (and hence, the local spin is a good quantum number) and perturbation theory is well defined in the original Anderson parameters. In graphene, where $\Delta(\omega) \propto \pi V^2 \rho(\omega)|\omega|^3/t^2$, $J(\mu) \approx V^2 U^2/[(\epsilon_0 - \mu)]$ is identically zero, the exchange coupling $|J| \sim V^2/\mu$ has no upper bound and can be shifted by gating towards the strong coupling limit of the Kondo problem, $J \rightarrow \infty$, when the Fermi level is tuned to the Dirac point, $\mu \rightarrow 0^{+}$. Since the experimentally accessible range of gate voltage for graphene on a 300 nm thick SiO$_2$ substrate is $\mu \in [-0.3, 0.3]$ eV, the exchange coupling of a magnetic Co adatom, for instance, with $V = 1$ eV and $\epsilon_0 = -0.4$ eV, can be tuned continuously in the range $|J| \in 1.4 - 10$ eV. This effect, which is allowed by the low DOS in graphene, brings the unprecedented experimental possibility of controlling the exchange coupling and switching magnetic adatoms between different Kondo coupling regimes in the proximity of a QCP, as we show in Fig. 2a.

Since the determinant of the exchange coupling matrix in Eq. (6), $\det[J_{\alpha\alpha'}]$, is identically zero, the exchange Hamiltonian is diagonalized into a new basis where one of the channels decouples from the bath. The eigenvalues in the new basis are $J_{\alpha,k,k'} = \sum_{\sigma} \Theta_{\alpha,k}^* \Theta_{\alpha,k'}$ and $J_{\nu} = 0$, and hence, the generic one-level exchange Hamiltonian maps into
the problem of a single channel Kondo Hamiltonian, \( \mathcal{H}_c = -2 \sum_{k,k'} J_{s,k,k'} \mathbf{s} \cdot \mathbf{s}_{k,k'} \), where \( \mathbf{s} \) is the itinerant spin, regardless of the implicit valley degeneracy.

In the one-level problem, the renormalization of the constant \( J \) due to the coupling of the local spin with the bath is given by: \( J' = J - 2N_s J \rho(D)(D/t)^n \delta D/D \), after integrating out the high energy modes with energy \( D \) at the bottom of the band, where \( N_s = 1, 2 \) describes the number of sublattices the adatom effectively hybridizes. Since a DOS in the form \( \rho(\omega) \propto |\omega|^{\eta} \) has a scaling dimension \( r \), where \( r = 1 \) in graphene, the restoration of the mean cut-off in the “poor man’s scaling” analysis requires an additional rescaling \( J' \rightarrow (D + \delta D)/D^{r + \eta} J' \), which results in the beta function

\[
\beta(J) = \frac{dJ}{d \ln D} = -(r + \eta)J - 2N_s J^2 \rho(D)(D/t)^n. \tag{8}
\]

The renormalization group (RG) flow leads to an intermediate coupling (IC) fixed point at \( J_c = -(r + \eta)t/|2N_s \rho(D)|D^n| \), which separates the weak and strong coupling sectors. For type I orbitals (ohmic bath), one recovers the usual IC fixed point \( J_c = -r/(2N_s \rho(D)) \), whereas for type II (super-ohmic bath, \( \eta = 2 \)) \( J_c \approx -3t^2/(2N_s D) \) in the Dirac case \( (r = 1) \). In graphene, this new fixed point describes a one-channel Kondo problem in the presence of an effective fermionic bath with DOS \( \rho \propto |\omega|^2 \), where \( \bar{r} \equiv r + \eta = 3 \). Since the tree level scaling dimension of the hybridization \( V \) in the Anderson model \((1 - \bar{r})/2 \), the case \( \bar{r} = 1 \) corresponds to an upper critical scaling dimension, above which \( (\bar{r} > 1) \) \( V \) is an irrelevant perturbation in the RG sense\[22\]. In this situation, fluctuations are not important near the QCP, and the critical exponents are expected to be mean-field like, in contrast with the marginal case \( (\bar{r} = 1) \), where mean field cannot be trusted\[22\].

The RG analysis derived from the exchange Hamiltonian\[20\] can be verified directly from the hybridization Hamiltonian\[2\]. In the large \( N \) limit near the critical regime, singly occupied level states are enforced at the mean field level through the constraint \( \lambda(\sum_{m} f_{\bar{m}} f_{\bar{m}} - 1) = 0 \)[21], with \( N = 2 \) in the spin 1/2 case. The minimization of the energy \( \partial \mathcal{H}_c/\partial \lambda = 0 \) gives \( \lambda = \frac{2}{N} \sum_{\bar{m}} \omega m(\omega) G^{0\text{R}}_{\bar{m}\bar{m}}(\omega) \), where \( n(\omega) = \frac{\Theta(\omega - \mu)}{T} + 1 \) is the Fermi distribution, \( T \) is the temperature, and \( G^{0\text{R}}_{\bar{m}\bar{m}}(\omega) = -\langle T[f_{\bar{m}}(\omega)^* f_{\bar{m}}^\dagger(\omega)] \rangle \) is the self-energy of the \( \bar{m} \)-electrons in the presence of the graphene bath, where \( \Delta(\omega) = -\text{Im} G^{0\text{R}}_{\bar{m}\bar{m}}(\omega) \) defines the level broadening and \( G^{0\text{R}}_{\bar{m}\bar{m}}(\omega) = \frac{\Theta(\omega - \alpha|\phi_\alpha| + i0^+)}{2\pi} - |\phi_\alpha|^2 G^{\alpha\alpha}_{\bar{m}\bar{m}}(\omega) \) is the self-energy of the \( \bar{m} \)-electrons in the presence of the Fermi liquid weak coupling regime, at \( J/K \ll 1 \), and the strong coupling regime, for \( |J| > |J_c| \approx (2/N_s)\mu eV \), in the regime \( T_K \ll (D/2)[1 - J_c/J(\mu + 3\mu^2/D^2)]^{1/3} \), as shown in Fig. 2b. At the critical coupling \( (J = J_c) \),

\[
T_K = (D/2)[1 - J_c/J(\mu + 3\mu^2/D^2)]^{1/3}, \tag{11}
\]

and the fingerprint of the QCP at \( \mu = 0 \) can be observed in the scaling of the QCP temperature with \( \mu \) in the vicinity of the QCP, at \( J \sim J_c \). This scaling can be measured in STM, where the signature of the Kondo effect is manifested in the form of a Kondo resonance in the DOS at the Fermi level, for \( T < T_K \).

In Fig. 2b, we numerically calculate the scaling of the Kondo temperature in tight-binding. For type II orbitals, the \( \nu = 1/3 \) exponent in the Kondo temperature, \( T_K \propto |J - J_c|^\nu \), found in the linear cone approximation persists above room temperature, up to \( T_K/t \sim 1 \) (red curves). In the more standard ohmic case, for spins on top carbon sites (black curve of the inset), the scaling is linear \( (\nu = 1) \) at the mean-field level.

Tracing the conduction electrons in the exchange Hamiltonian\[20\], the RKKY Hamiltonian of a spin lattice in graphene is \( H_{RKKY} = -J^2 \sum_{i,j} \chi_{i,j}^x \mathbf{s}_i \cdot \mathbf{s}_j \), where
In the case of carbon nanotubes, where the RKKY interaction is strong, the decay is described by a fast power law $\alpha R^{-\gamma}$, where $\alpha$ and $\gamma$ are constants. The inset of Fig. 3 shows the decay of the magnetic susceptibility $\chi(R)$ vs distance $R$, along a zigzag direction (in lattice units), for $A$-sites (blue squares), $H$-sites (red circles), and $S$ sites for spins on the same sublattice (red circles). Solid lines: $\mu = 0$; dashed: $\mu = t$. Inset: $\chi_{ij}$ plot in a log scale. Orange (light) guide line: $1/R^3$; black: $1/R^7$. On the right: $\chi(q)$ for $A$ site spins, plotted in the graphene BZ at b) $\mu = 0$ and c) $\mu = t$. Red (dark) regions represent $\chi(q) > 0$ and blue (light) regions $\chi(q) < 0$.

$$\chi_{ij}^{xy}(R_i - R_j) = \chi^{xy}(R_i - R_j)$$ is the spin susceptibility, with $i, j$ indexing the local spins, and $x, y = A, B, H, S_A, S_B$ labeling the position of the magnetic adatoms in lattice. In momentum space,

$$\chi^{xy}(q) = \sum_{\alpha, \alpha', \mu, \nu, q} \mathcal{M}^{xy}_{\alpha, \alpha', \mu, \nu, q} \frac{n[E_{\nu}(p + q) - n[E_{\nu}(p) - E_{\nu}(p + q)]}{1 + \Theta_{\alpha, \alpha'}^{xy}(p + q)} ,$$

(12)

where $\mathcal{M}^{xy}_{\alpha, \alpha', \mu, \nu, q} = \Theta_{\alpha, \alpha'}^{xy}(p + q) \Theta_{\alpha, \alpha'}^{xy}(p + q) \Theta_{\alpha, \alpha'}^{xy}(p + q) / |\phi_{\nu}(p + q)|$, in agreement with Ref. [25], in the Dirac cone limit. For an $H$-site [24],

$$\mathcal{M}^{HH} = |\Theta_{\alpha, \alpha'}^{H}(p + q)|^2 ,$$

(13)

where $|\Theta_{\alpha, \alpha'}^{H}(p + q)|^2 = |\phi_{\nu}(p + q)|^2 [1 + (-1)^\alpha \text{Re}(\Theta_{\alpha, \alpha'}^{H}(p + q))]^2$ for orbitals of type II; for $S$-sites, $\mathcal{M}^{SA}$ for spins on the same sublattice and $\mathcal{M}^{SB}$ for opposite sublattices [24] [25].

In Fig. 3a, we show the spatial decay of the RKKY interaction on the lattice for type II orbitals on $A$, $H$ and $S_A$ site spins. At half filling, the RKKY interaction is always ferromagnetic for same sublattice spins, substitutional or not, and antiferro for spins on opposite sublattices [24] [25]. The $H$ case on the other hand, is ferromagnetic for nearest neighbor spins and antiferromagnetic at longer distances (blue squares). In the $H$ and $S$ cases, the interaction is short range and decays with a fast power law $\sim 1/R^3$, in contrast to the known $1/R^5$ decay in the $A$ site case [24] [25] [27] [28], as shown in the inset of Fig. 3. This fast decay is consistent with the case of carbon nanotubes, where the RKKY interaction decays with $1/R$ for top carbon sites and with $1/R^2$ for isotropic orbitals on $H$ sites [24].

Fig. 3b and 3c display the magnetic peaks in the susceptibility in the $A$-site case for $\mu = 0$, and $\mu = t$. For $\mu < t$, $\chi(q)$ has a strong ferromagnetic forward scattering peak around the center of the BZ ($q = 0$), and six sub-dominant antiferromagnetic peaks at corners of the BZ. Exactly at $\mu = t$, a strong peak emerges at the $M$ point due to the nesting of the Van-Hove singularities (VHS) of the graphene band (see Fig. 3c), where the DOS diverges logarithmically. This peak reverses the ordering pattern of the RKKY interaction in comparison to the $\mu = 0$ regime in all studied cases, as shown in the dashed lines of Fig. 3a. When $\mu$ is at the VHS, the interaction between spins on same (opposite) sublattices, substitutional or not, is always antiferromagnetic (ferro). In the same way, the RKKY interaction in the $H$ site case becomes antiferromagnetic for nearest neighbor sites and ferromagnetic at long distances.

In conclusion, we have derived the one-level-exchange Hamiltonian for magnetic adatoms in graphene and shown the existence of two symmetry classes of magnetic orbitals that correspond to distinct classes of Kondo QCP. We also showed that the exchange coupling can be controlled across the quantum critical region with the application of a gate voltage.

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In Ref. [1] we have predicted the existence of two different classes of quantum critical points for the Kondo problem in graphene, which was shown to correspond effectively to the problem of a localized spin 1/2 coupled to a fermionic bath with electronic density of states $\rho(\omega) \propto |\omega|^{\bar{r}}$, with $\bar{r} = 1$ or 3. Here we point out that the mean field exponent $\nu = 1/3$ derived within the slave boson approach for the class of orbitals of type II ($\bar{r} = 3$) is incorrect.

Above the upper critical scaling dimension of the Anderson model ($\bar{r} > 1$), the intermediate coupling fixed point is non-interacting and describes the level crossing between singlet and doublet states with the trivial exponent $\nu = 1$. Albeit fluctuations do not play a role in the critical behavior for $\bar{r} > 1$, the critical theory is not of the Ginzburg-Landau type and the validity of the mean-field slave boson equation of state (9) breaks down in the $\bar{r} = 3$ class, invalidating Eq. (10), (11), and the inset of Fig. 2b for the case of type II orbitals.

All the other results of the paper remain valid, including the spin exchange Hamiltonian in Eq. (6) and the prediction of a fast power law for the spatial decay of the RKKY interaction for type II orbitals ($\sim 1/R^7$).

We note that since hyperscaling is not obeyed for $\bar{r} > 1$, the scaling prediction for the Kondo temperature with the chemical potential, $\mu$, in the quantum critical region, $T_K \propto |\mu|$, can be violated[3]. In the situation where the scaling prediction fails, the criticality in the $\bar{r} = 1$ and $\bar{r} = 3$ classes can be in principle distinguished. That will be verified with NRG methods elsewhere.

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