Reentrant Kondo effect in Landau-quantized graphene: Influence of the chemical potential

Balázs Dóra*
Max-Planck-Institut für Physik Komplexer Systeme, Nöthnitzer Strasse 38, 01187 Dresden, Germany

Peter Thalmeier
Max-Planck-Institut für Chemische Physik fester Stoffe, 01187 Dresden, Germany

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We have studied the effect of an Anderson impurity in Landau quantized graphene, with special emphasis on the influence of the chemical potential. Within the slave-boson mean-field theory, we found reentrant Kondo behavior by varying the chemical potential or gate voltage. Between Landau levels, the density of states is suppressed, and by changing the graphene's Fermi energy, we cross from metallic to semiconducting regions. Hence, the corresponding Kondo behavior is also influenced. The f-level spectral function reveals both the presence of Landau levels in the conduction band and the Kondo resonance.

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I. INTRODUCTION

The existence of Dirac fermionic excitations in single layer graphene has attracted great interest.[1–6] Theoretically, the linear dispersion and chiral nature of massless quasiparticles has many unexpected physical consequences, especially for magnetotransport properties.[7–11] Experimentally, graphene may be considered as a zero gap semiconductor with charge carriers of very high mobility whose density may easily be controlled by applying a gate voltage, which gives hope for interesting applications.[6]

The scattering of graphene quasiparticles from normal impurities determines electronic and thermal transport and is well studied.[7,9–13] The linear dispersion of quasiparticles and the associated pseudogap plays an essential role in the magnetotransport[11] because it leads to unconventional Landau level quantization as compared to the case of parabolic bands. One essential difference is that a zeroth order Landau level is pinned at zero energy for any field strength. With increasing impurity scattering, the pseudogap is therefore gradually filled and the density of states exhibits oscillations as a function of the chemical potential or carrier number.[9,11]

Because the graphene carrier number and density of states can easily be manipulated with gate voltage and magnetic field, it is attractive to consider the effect of magnetic impurities in a graphene host. It is well known that magnetic moments in the Fermi sea show the competition of on-site Kondo singlet formation and intersite RKKY interactions, which is highly sensitive to the density of states around the Fermi level.[14] While experiments in graphene are still missing, we think that chemisorption or adsorption of transition metal ions on graphene sheets is the most likely realization of Kondo effect and RKKY interaction with Dirac electrons. Possible ways for creation of local moments in carbon based materials have been discussed in Refs. 15–17.

Theoretically, the Kondo- and Anderson-type models for local moments in graphene have been studied without magnetic field.[16,18] In the present work, we investigate the effect of Landau level formation on the screening of magnetic impurities described by the Anderson model. Due to the strong density of state variation with chemical potential and magnetic field, it should be possible to drive the magnetic impurity in and out of the Kondo regime, which is characterized by the formation of an f-level resonance close to the chemical potential. This effect is studied within the mean-field slave-boson theory and assuming a constant broadening of Landau levels by normal impurities. In Sec. II, we define the Anderson impurity model coupled to a Dirac Fermi sea in a magnetic field. The solution of the model within the saddle-point approximation is derived in Sec. III and the numerical results are discussed in Sec. IV. Finally, Sec. V gives the conclusion.

II. ANDERSON IMPURITY IN GRAPHENE

The Hamiltonian of Dirac quasiparticles living on a single graphene sheet, interacting with an infinite-U Anderson impurity at the origin, is given by[7,19,20]

$$H_0 = \sum_{\sigma=\pm1}\left(\int d\mathbf{r}\sum_{s=\pm1}\Psi^+_{\sigma,s}(\mathbf{r})\left\{\sum_{j=x,y} -v_F(1-2\delta_{j,y}\delta_{s,-1})\times\sigma_j[-i\partial_j + eA_j(\mathbf{r})] - \mu - h\sigma\right\}\Psi_{\sigma,s}(\mathbf{r}) + (E-h\sigma)f^+_\sigma f_\sigma + V\sum_{s=\pm1}[f^+_\sigma\Psi_{\sigma,s}(\mathbf{0})b + b^+\Psi^+_{\sigma,s}(\mathbf{0})f_\sigma]\right), \quad (1)$$

where σ_j's are the Pauli matrices and stand for Bloch states residing on the two different sublattices of the bipartite hexagonal lattice of graphene.[7,9] The quasiparticle spectrum vanishes at six points in the Brillouin zone. Out of these six, only two are nonequivalent points and are referred to as K and K' points in the Brillouin zone, denoted by the $s=1$ and -1 indices, respectively, and δ is the Kronecker delta. The Kronecker delta function accounts for the nonequivalence of the two Dirac points[8] at K and K'. The finite chemical potential μ accounts for particle-hole symmetry breaking. The vector potential for a constant, arbitrarily oriented magnetic field reads as $\mathbf{A}(\mathbf{r})=[-By\cos\theta, 0, B(y\sin\theta\cos\phi - x\sin\theta\sin\phi)]$, where θ is the angle the magnetic field makes from the z axis and ϕ is the in-plane azimuthal angle measured from the x axis. The Zeeman term is assumed to couple to both the impurity and Dirac electrons by the same

g factor, $h=g\mu_B B$ and $v_F \approx 10^6$ m/s, and is characteristic to graphene. f_σ^+ and f_σ creates and annihilates an electron on the localized E level; b^+ and b are the slave-boson operators, responsible for the hole states.[21–23] These take the infinite-U term into account.

The Hamiltonian should be restricted to the subspace

$$\sum_{\sigma=\pm 1} f_\sigma^+ f_\sigma + b^+ b = 1. \tag{2}$$

Within the mean-field approximation, the slave-boson operators are replaced by their expectation value, $b_0=\langle b\rangle$, and the constraint is satisfied by introducing a Lagrange multiplier λ,

$$H = \sum_{\sigma=\pm 1}\left(\int d\mathbf{r} \sum_{s=\pm 1} \Psi_{\sigma,s}^+(\mathbf{r})\left\{\sum_{j=x,y} -v_F(1-2\delta_{j,y}\delta_{s,-1}) \times \sigma_j[-i\partial_j + eA_j(\mathbf{r})] - \mu - h\sigma\right\}\Psi_{\sigma,s}(\mathbf{r}) + (E+\lambda-h\sigma)f_\sigma^+ f_\sigma + Vb_0 \sum_{s=\pm 1}[f_\sigma^+ \Psi_{\sigma,s}(\mathbf{0}) + \Psi_{\sigma,s}^+(\mathbf{0}) f_\sigma]\right) + \lambda(b_0^2 - 1). \tag{3}$$

In the particle-hole symmetric case ($\mu=0$), in the absence of magnetic field, the energy spectrum of the system of Dirac electrons is given by

$$E(\mathbf{k}) = \pm v_F|\mathbf{k}|. \tag{4}$$

This describes massless relativistic fermions with a spectrum consisting of two cones, touching each other at the end points. From this, the density of states per spin follows as

$$\rho(\omega) = \frac{1}{\pi}\sum_{\mathbf{k}} \delta[\omega - E(\mathbf{k})] = \frac{1}{\pi}\frac{A_c}{2\pi}\int_0^{k_c} k dk \delta(\omega \pm v_F k) = \frac{2|\omega|}{D^2}, \tag{5}$$

where k_c is the cutoff, $D=v_F k_c$ is the bandwidth, and $A_c = 4\pi/k_c^2$ is the area of the hexagonal unit cell. We mention in passing that an applied gate voltage directly controls the number of extra charge carriers in the system, which is given by $eV \sim n = \int_0^\mu \rho(\omega) d\omega = \mu^2/D^2$. Hence, chemical potential is proportional to the square root of the applied gate voltage even in the ideal case, without any scatterers and magnetic field. Such a relation can hardly be calculated for the realistic case. Nevertheless, the chemical potential always varies monotonically with the gate voltage due to the positiveness of the density of states.

Magnetic impurities in gapless Fermi systems have thoroughly been studied starting with the pioneering work of Withoff and Fradkin,[24] and the focus was on the influence of gapless excitations on the Kondo phenomenon (for a review, see Ref. 25). Recently, the Kondo effect in graphene without magnetic field has been studied within this framework.[16] Here, we allow for Landau quantization of the quasiparticle spectrum and study the orbital effect of magnetic field on the various Kondo phases.

In the presence of magnetic field, the eigenvalue problem of our Hamiltonian without the localized level can readily be solved.[7] From now on, we concentrate on the K point; the eigenfunctions of the K' point can be obtained by exchanging the two components of the spinor. Momentarily, we also neglect the spin and the Zeeman term and concentrate on the effect of Landau quantization. They will be reinserted when necessary. For the zero energy mode ($E=0$), the eigenfunction is obtained as

$$\Psi_k(\mathbf{r}) = \frac{e^{ikx}}{\sqrt{L}}\begin{pmatrix} 0 \\ \phi_0(y - kl_B^2)\end{pmatrix}, \tag{6}$$

and the two components of the spinor describe the two bands. The energy of the other modes reads as

$$E(n,\alpha) = \alpha\omega_c\sqrt{n+1}, \tag{7}$$

with $\alpha=\pm 1$, $n=0,1,2,\ldots$, and $\omega_c = v_F\sqrt{2e|B\cos(\theta)|}$ is the Landau scale or energy but is different from the cyclotron frequency.[26] Only the perpendicular component of the field enters into these expressions, and by tilting the field away from the perpendicular direction corresponds to a smaller effective field. The sum over integer n's is cut off at N given by $N+1=(D/\omega_c)^2$, which means that we consider $2N+3$ Landau levels altogether.

The corresponding wave function is

$$\Psi_{n,k,\alpha}(\mathbf{r}) = \frac{e^{ikx}}{\sqrt{2L}}\begin{pmatrix} \phi_n(y - kl_B^2) \\ \alpha\phi_{n+1}(y - kl_B^2)\end{pmatrix}, \tag{8}$$

with cyclotron length $l_b = 1/\sqrt{eB}$. Here, $\phi_n(x)$ is the nth eigenfunction of the usual one-dimensional harmonic oscillator. The electron-field operator at the K point can be built up from these functions as

$$\Psi(\mathbf{r}) = \sum_k \left[\Psi_k(\mathbf{r}) c_k + \sum_{n=0}^{N}\sum_{\alpha=\pm 1} \Psi_{n,k,\alpha} c_{k,n,\alpha}\right]. \tag{9}$$

The Green's functions of these new operators do not depend on k and read as

$$G_0(i\omega_n, k) = \frac{1}{i\omega_n}, \tag{10}$$

$$G_0(i\omega_n, k, n, \alpha) = \frac{1}{i\omega_n - E(n,\alpha)}, \tag{11}$$

for c_k and $c_{k,n,\alpha}$, respectively, and ω_n is the fermionic Matsubara frequency. As seen from above, the density of states in the presence of quantizing magnetic field contains Dirac-delta peaks located at the Landau level energies, and quasiparticle excitations have infinite lifetime. To describe a more realistic situation, scattering from disorder needs to be considered in the presence of the magnetic field. Usually, the resulting self-energy of the Dirac fermions, determined in a self-consistent manner, depends on the frequency and field strength.[7,11] However, good agreement can be reached by assuming a constant, phenomenological scattering rate, denoted by Γ, for small and moderate fields, as can be learned from similar analysis.[9,27] To simplify calculations, we have chosen to mimic disorder by a constant scattering rate.

III. SADDLE-POINT EQUATIONS

The free energy of the system can be found from Eq. (3) using a standard technique.[23] The value of λ and b_0 is determined self-consistently by minimizing the free energy of the system with respect to them.[28,29] As a result, by restoring the finite chemical potential, spin, and the Zeeman term, the saddle-point equations at $T=0$ are given by

$$b_0^2 = \sum_{\sigma=\pm 1} S_1(E+\lambda-\sigma h, \mu+\sigma h), \tag{12}$$

$$\lambda = \sum_{\sigma=\pm 1} S_2(E+\lambda-\sigma h, \mu+\sigma h), \tag{13}$$

where the auxiliary functions are defined as

$$S_1(y,z) = \int_0^\infty \frac{dx}{\pi} \frac{y + b_0^2 2V^2 \,\mathrm{Im}\,\Sigma(x+\Gamma-iz)}{[x + b_0^2 2V^2 \,\mathrm{Re}\,\Sigma(x+\Gamma-iz)]^2 + [y + b_0^2 2V^2 \,\mathrm{Im}\,\Sigma(x+\Gamma-iz)]^2}, \tag{14}$$

$$S_2(y,z) = 2V^2 \int_0^\infty \frac{dx}{\pi} \frac{[x + b_0^2 2V^2 \,\mathrm{Re}\,\Sigma(x+\Gamma-iz)]\mathrm{Re}\,\Sigma(x+\Gamma-iz) + \mathrm{Im}\,\Sigma(x+\Gamma-iz)[y + b_0^2 2V^2 \,\mathrm{Im}\,\Sigma(x+\Gamma-iz)]}{[x + b_0^2 2V^2 \,\mathrm{Re}\,\Sigma(x+\Gamma-iz)]^2 + [y + b_0^2 2V^2 \,\mathrm{Im}\,\Sigma(x+\Gamma-iz)]^2}, \tag{15}$$

$$\Sigma(z) = \frac{1}{N+1}\left[\frac{1}{z} + \sum_{k=0}^{N} \frac{2z}{z^2 + \omega_c^2(k+1)}\right] = \frac{1}{N+1}\left\{\frac{1}{z} + 2\frac{z}{\omega_c^2}\left[\Psi\left(\frac{z^2}{\omega_c^2} + N + 2\right) - \Psi\left(\frac{z^2}{\omega_c^2} + 1\right)\right]\right\}. \tag{16}$$

The $1/(N+1) \propto B$ prefactor denotes the Landau level degeneracy. These reduce to the standard saddle-point equations for gapless phases[28,30] for zero field and $\Gamma=0$. The extra factor of 2 in front of V^2 stems from the two nonequivalent Dirac cones at the K and K' points since by using

$$\int_{-\infty}^{\infty} dk \phi_n(y - k l_B^2)\phi_m(y - k l_B^2) = \frac{\delta_{n,m}}{l_B^2}, \tag{17}$$

each Landau level at each valley contributes to the hybridization energy by V^2. Equation (12) accounts for the constraint of having at most one f electron at the impurity site [Eq. (2)]. Equation (13) stems from the equation of motion of the slave boson-field b: since it is constant in the mean-field approach, the terms determining its dynamics should add up to zero.[22] Here, $\Sigma(z)$, which is related to the f-level self-energy, contains all the information about the conduction electron bath, where the magnetic impurity is embedded. When the field strength goes to zero, these equations reduce to those found in gapless phases[28] as

$$\Sigma(z) = 2\frac{z}{D^2}\ln\left(1 + \frac{D^2}{z^2}\right). \tag{18}$$

In this case, for $\mu=\Gamma=0$, the critical f-level energy is found to be $E_c=-8V^2/D$. For $E<E_c$, only the trivial solution occurs ($b_0=0$); hence, charge fluctuations are completely suppressed. The solution of Eqs. (12) and (13) can be carried out by realizing that Eq. (12) depends only on the renormalized f-level energy $E+\lambda$ and not separately on the two variables E and λ. Then, by fixing the value of $E+\lambda$, we can determine the corresponding b_0 by iteration, for example. By inserting the values of the renormalized f-level energy and the slave-boson expectation value to Eq. (13), we can directly read off λ and determine E by subtracting it from the renormalized f-level energy. As in other similar problems, this method predicts a quantum phase transition at $T=0$, characteristic to gapless Kondo phases. However, in our case, the order of the transition can change from second to first. Such a transition is absent for magnetic impurities embedded to normal metals.[23]

IV. SPECTRAL FUNCTION: DISCUSSION

The full f-electron spectral function per spin along the real frequency axis reads as

$$\rho_{f,\sigma}(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\,\frac{b_0^2}{\omega - E - \lambda + \sigma h - 2V^2 b_0^2 \Sigma_f(\omega + \mu + \sigma h + i\Gamma)}, \tag{19}$$

where $\Sigma_f(x) = -i\Sigma(-ix)$. It shows the Landau level oscillations and, in addition, a large Kondo peak whenever a nontrivial solution of Eqs. (12) and (13) exists. Without magnetic field and normal impurities, it shows a marginal Fermi liquid behavior,[31] as can be observed from the analytically continued f-level self-energy,

$$\Sigma_f(\omega \ll D) \approx -\frac{4V^2 b_0^2}{D^2}\left[2\omega \ln\left(\frac{D}{|\omega|}\right) + i\pi|\omega|\right], \tag{20}$$

which, in the presence of normal impurities, turns into the usual fermionic self-energy at low energies, with ω being replaced by Γ, the scattering rate. In the case of quantizing magnetic field, without normal impurities, its imaginary part consists of Dirac-delta functions at the Landau level energies. Nonmagnetic impurities smear these singularities by transforming them into Lorentz functions. Hence, the self-

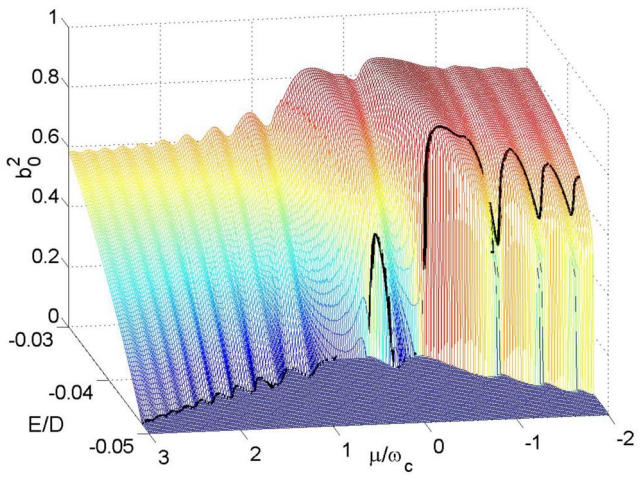

FIG. 1. (Color online) The order parameter is shown for $N=1000$, $2(V/D)^2=0.01$, $\Gamma=0.05\omega_c$, $h=0.1\omega_c$. For negative chemical potentials, the order of the transition changes from second to first, as is denoted by the thick black line.

energy is that of a well-formed Fermi liquid for realistic situations. We mention the possibility of detecting non-Fermi liquid phases[15,16] in graphene due to the valley degeneracy, which might originate from multichannel Kondo physics. This can be signaled by the finite critical value of E even at $T=0$, as is seen in Fig. 1. However, such a situation is unlikely to be observed by our simple mean-field analysis.

The numerical solution of the saddle-point equations has been carried out, and the result for the order parameter (b_0^2) is shown in Fig. 1. It is directly related to the f-level occupation through Eq. (2) as $n_f=1-b_0^2$. In general, by increasing $|\mu|$, a Fermi surface develops, and the critical value of E penetrates deeply into the $E<0$ region.[24] However, there is a crucial difference between positive and negative chemical potentials: for the former, the Kondo energy scale becomes extremely small, as can be seen in Fig. 2 in the f-level density of states, since the f-level energy is well below the Fermi energy of conduction electrons. In the latter case, the energy level of f electrons lies above the Fermi energy; hence, the Kondo scale enhances.

When the chemical potential is close to a Landau level energy $E(n,\alpha)$, the nontrivial solution (nonzero b_0) extends further in the $E<0$ region because the density of states is enhanced around Landau level energies. Between Landau levels, the conduction electron density of states is suppressed, and the critical f-level energy moves closer to zero, leading to the oscillatory behavior in the phase boundary and diagram, as can be seen in Figs. 1 and 2. Here, we assume that $N=1000$, which corresponds to weak or moderate fields,[11] depending on the explicit value of the cutoff D. The Zeeman term is chosen to be much smaller than ω_c and follows from actual numbers in graphene.[9] As a result, by changing the chemical potential, we can move between Landau levels and we can enter into and leave the Kondo regime. Hence, reentrant behavior is found. By decreasing the scattering rate Γ from normal impurities, the oscillations along the phase boundary become more pronounced.

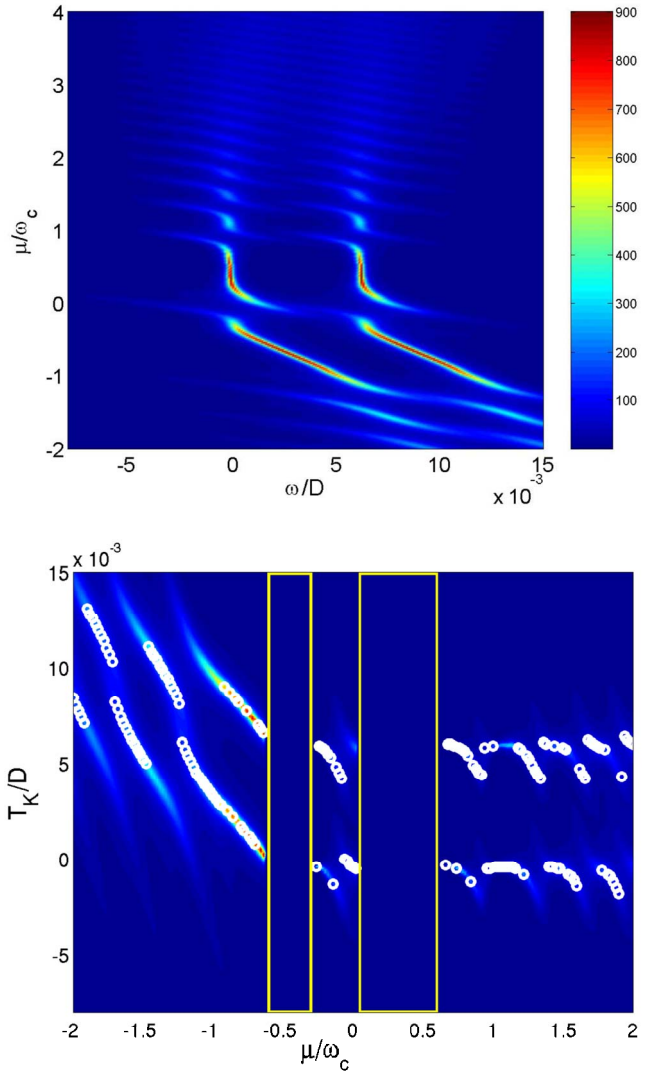

FIG. 2. (Color online) The contour plot (for $E/D=-0.036$, upper panel) of the f-level density of states and the Kondo temperature (T_K for $E/D=-0.0387$, lower panel) are shown for $N=1000$, $2(V/D)^2=0.01$, $\Gamma=0.05\omega_c$, $h=0.1\omega_c$. The parallel structures in the contour plot denote the Kondo peaks, separated by twice the Zeeman energy. The reentrant behavior is clearly observable as a function of the chemical potential in the lower panel, where the Kondo peaks are emphasized by the bright circles. The yellow (bright) framed bars denote regions without Kondo physics ($b_0=0$).

The reentrant behavior can more directly be checked in the f-level density of states in Fig. 3, where large Kondo peaks are observable (due to Zeeman splitting) in addition to a small oscillation stemming from Landau levels in the conduction band, when the nontrivial solution exists. The distance between the two parallel ridges in the contour plot in Fig. 2 is twice the Zeeman energy, as it should be. When only the trivial solution exists ($b_0=0$), $\rho_f(\omega)$ is completely suppressed since the maximally allowed one particle always occupies the f level.

In the Kondo regime, as one varies the chemical potential, the Kondo temperature does not change monotonically. It remains mainly pinned to the closest Landau level, and then

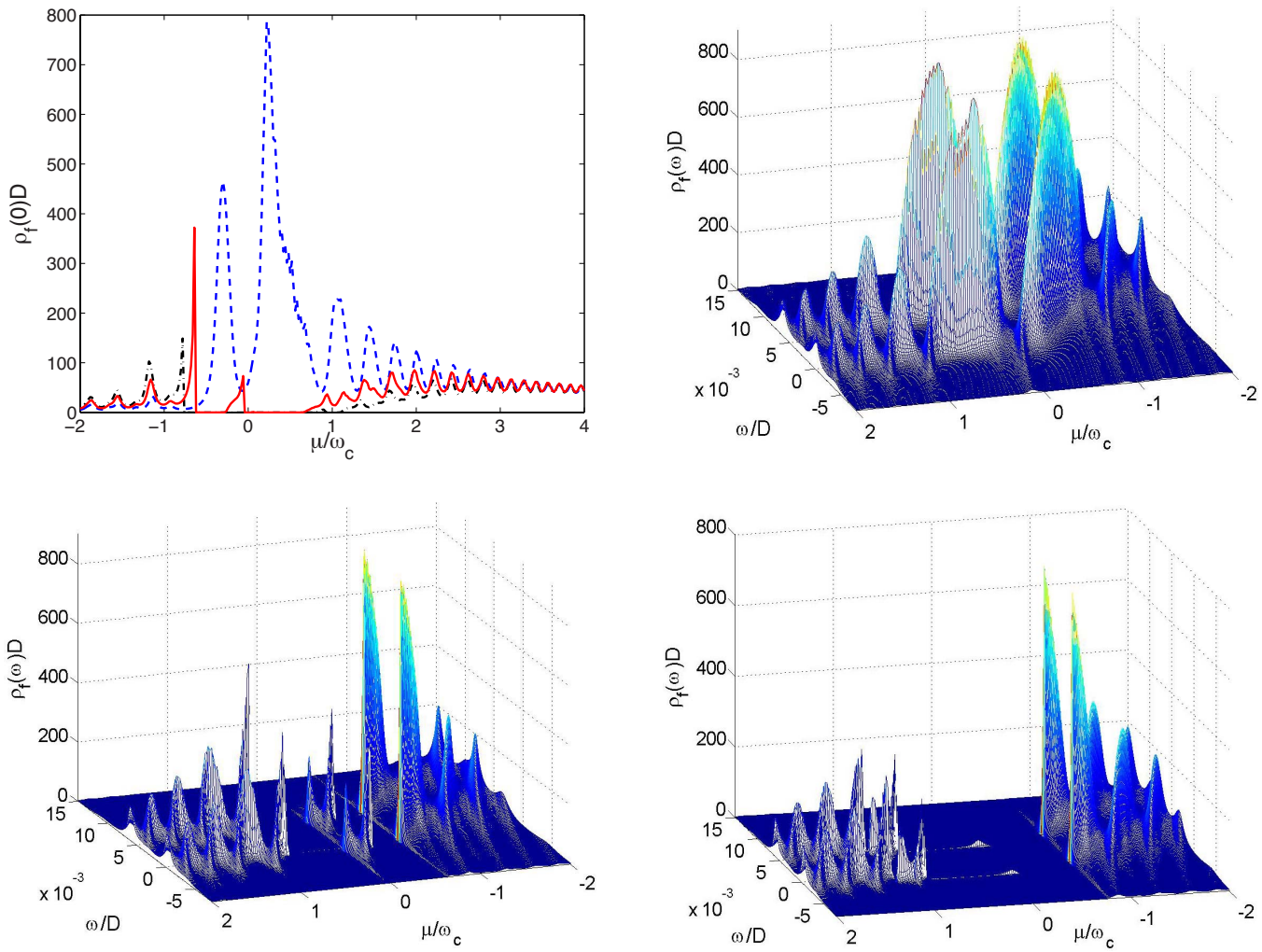

FIG. 3. (Color online) In the top left panel, the spin averaged f-level density of states is shown as a function of the chemical potential for $N=1000$, $2(V/D)^2=0.01$, $\Gamma=0.05\omega_c$, $h=0.1\omega_c$ for $E/D=-0.036$ (blue dashed), -0.0387 (red solid), and -0.04 (black dashed dotted). Note the presence of states for the middle value close to zero chemical potential. Reentrant behavior is also observable close to the first Landau level for $E/D=-0.04$, $\mu\approx\omega_c$. The three-dimensional plots show the evolution of the spectral density as a function of chemical potential and frequency for the previous three E values from top to bottom, left to right. Note the presence of small islands of states for $E/D=-0.0387$, responsible for the Kondo effect at $\mu\approx 0$.

suddenly jumps to the neighboring one, as is seen in Fig. 2.

These features in the density of states can probably be detected by conductance measurements, which measure directly the inverse of the f-level density of states, in addition to normal impurities.[23] When $\rho_f(0)$ is finite, its contribution is thought to overwhelm that of normal impurities.[23] Then, both the Landau level oscillations stemming from the orbital quantization of conduction electrons and the Kondo behavior could be seen. The change in the Kondo temperature (the peak position in Figs. 2 and 3) by chemical potential or gate voltage can also, in principle, be detected from the specific heat. This is expected to exhibit a double peak structure around T_K due to the Zeeman term. However, such measurements on thin graphene films are extremely demanding. The change of the spectral function would reveal itself directly in photoemission spectroscopy,[4,5] which, however, in a finite magnetic field, does not constitute a standard experiment.

The presence of Kondo resonance makes itself felt in magnetic responses, which are expected to be different from the usual Kondo behavior due to the presence of orbital (or diamagnetic) terms. This already influences the magnetic susceptibility of pure graphene and probably alters the magnetic behavior of the impurity as well.

V. CONCLUSIONS

In summary, we have studied the infinite-U Anderson impurity embedded to a host of two-dimensional Dirac fermions within the self-consistent slave-boson mean-field theory. The host material corresponds to graphene, where the elementary excitations on a honeycomb lattice are Dirac fermions. Such a system can most probably be realized by the chemisorption of transition metal ions on graphene sheets. We allow for the Landau quantization of the conduction electron spectrum, which turns out to be unusual[7,32] ($E\propto\sqrt{n}$) in comparison with normal metals ($E\propto n$). Then, we study the effect of *orbitally quantizing magnetic field* on the Kondo phenomenon, in addition to the Zeeman term, albeit the latter

is thought to be negligible in the presence of Landau levels.[9]

When the chemical potential lies close to a Landau level energy, the conduction electron density of states is enhanced, and the mixed-valence regime extends further in the $E<0$ region.[24] Between Landau levels, the density of states resembles to that in an insulator, and the local moment regime gains ground. Hence, by varying the chemical potential between Landau levels, reentrant Kondo behavior is found. This manifests itself in the f-electron spectral function, which accommodates small islands of states, corresponding to the mixed-valence case, separated by the deserts of local moment regions, as μ changes. The reentrant behavior manifest itself strongly around the zeroth order Landau level, but for clean samples, it should be observable at higher levels as well. The experimental detection of this phenomenon can be done by conductance measurements in magnetic field at low temperatures, which can show the presence or absence of Kondo peaks in the f-electron density of states.

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*dora@kapica.phy.bme.hu

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