

# Free Magnetic Moments in Disordered Metals

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The screening of magnetic moments in metals, the Kondo effect, is found to be quenched with a finite probability in the presence of nonmagnetic disorder. Numerical results for a disordered electron system show that the distribution of Kondo temperatures deviates strongly from the result expected from random matrix theory. A pronounced second peak emerges for small Kondo temperatures, showing that the probability that magnetic moments remain unscreened at low temperatures increases with disorder. Analytical calculations, taking into account correlations between eigenfunction intensities yield a finite width for the distribution in the thermodynamic limit. Experimental consequences for disordered mesoscopic metals are discussed.

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In a metal with antiferromagnetic exchange interaction between a local magnetic moment and the conduction electrons, correlations cause a change in the Fermi liquid ground state. The screening of the localized spin by the formation of a Kondo singlet below the Kondo temperature,  $T_K$ , is affected by disorder in various ways. Fluctuations in the exchange coupling due to random positioning of magnetic moments results in a dispersion of  $T_K$  [1]. Since  $T_K$  is defined by an integral equation similar to the BCS equation for the critical temperature of a superconductor, one could expect, by analogy, the Anderson theorem [2] to be valid. In that case, the leading correction to  $T_K$  would be of order  $T_K^{(0)}/g$ , where  $T_K^{(0)}$  is the bulk Kondo temperature in the clean limit and  $g = E_F\tau$ , the dimensionless conductance, is the ratio between the Fermi energy  $E_F$  and the elastic scattering rate  $1/\tau$ . However, the equation for  $T_K$  requires a sum over the local density of states (LDOS) rather than the global one (DOS). It is known that disorder induces correlations in energy in the LDOS [3]. Thus, impurities cause additional fluctuations on the LDOS and thereby on  $T_K$ .

Let us begin by describing the relevant approaches to the different regimes in the problem. Consider a single magnetic impurity in a closed, phase-coherent metallic grain where the energy levels are discrete, a Kondo box [4, 5], with mean level spacing  $\Delta = 1/\nu L^d$  and Thouless energy  $E_c = D_e/L^2$ . Here,  $L$  is the system linear size,  $\nu$  the DOS,  $D_e$  the diffusion constant, and  $d$  the space dimension. We note that  $T_K$  does not exceed  $1/\tau$  in metals, ruling out perturbation theory in  $1/\tau$ . However, the regime  $E_c < T_K < 1/\tau$  applies to a wide range of

metals and an expansion in terms of diffusion diagrams is permitted in that case [6]. For smaller grains, when  $\Delta < T_K < E_c$ , random matrix theory (RMT) yields a distribution of  $T_K$  that scales with  $\Delta$  alone. When the grain is so large that the localization length  $\xi$  is smaller than  $L$ ,  $E_c$  and  $\Delta$  are irrelevant and  $T_K$  is determined by the average energy level spacing in the vicinity of the magnetic impurity,  $\Delta_\xi = 1/\nu \xi^d$  [7]. Thus, one expects RMT to be applicable on the scale  $\Delta_\xi$  in this local random matrix theory (LRMT) regime. However, corrections to RMT and LRMT due to correlations of wave functions of order  $1/g$  are found to be essential for determining  $T_K$ . Finally, when  $T_K$  is smaller than  $\Delta$  or  $\Delta_\xi$ , the distribution of  $T_K$  is mainly determined by the coupling of the magnetic impurity to a two-level system.

In the symmetric Anderson model of a magnetic impurity [8], the exchange interaction matrix elements are estimated as  $J_{kl} = 4t_{ik}^*t_{il}/U$ , where  $U$  is the on-site Coulomb energy and  $t_{ik}$  the hopping matrix elements connecting the localized state  $\phi_i(\mathbf{r})$  with a delocalized state  $\psi_k(\mathbf{r})$ :  $t_{ik} = \int d^d r \phi_i^*(\mathbf{r}) \hat{T} \psi_k(\mathbf{r})$ , where  $\hat{T}$  is the kinetic energy operator. For states localized at  $\mathbf{r} = 0$  with a radius  $a_0$ ,  $J_{kl} \approx J \psi_k^*(0) \psi_l(0)$ , where  $J = 4(1/m^* a_0^2)^2/U$  is the exchange coupling,  $m^*$  the band mass. In the antiferromagnetic case ( $J < 0$ ),  $J$  is strongly renormalized. The temperature at which the second-order correction is equal to the bare value defines  $T_K$  [9],

$$1 = \frac{1}{2x} \sum_{n=1}^N \frac{x_n}{s_n} \tanh\left(\frac{s_n}{2\kappa}\right), \quad (1)$$

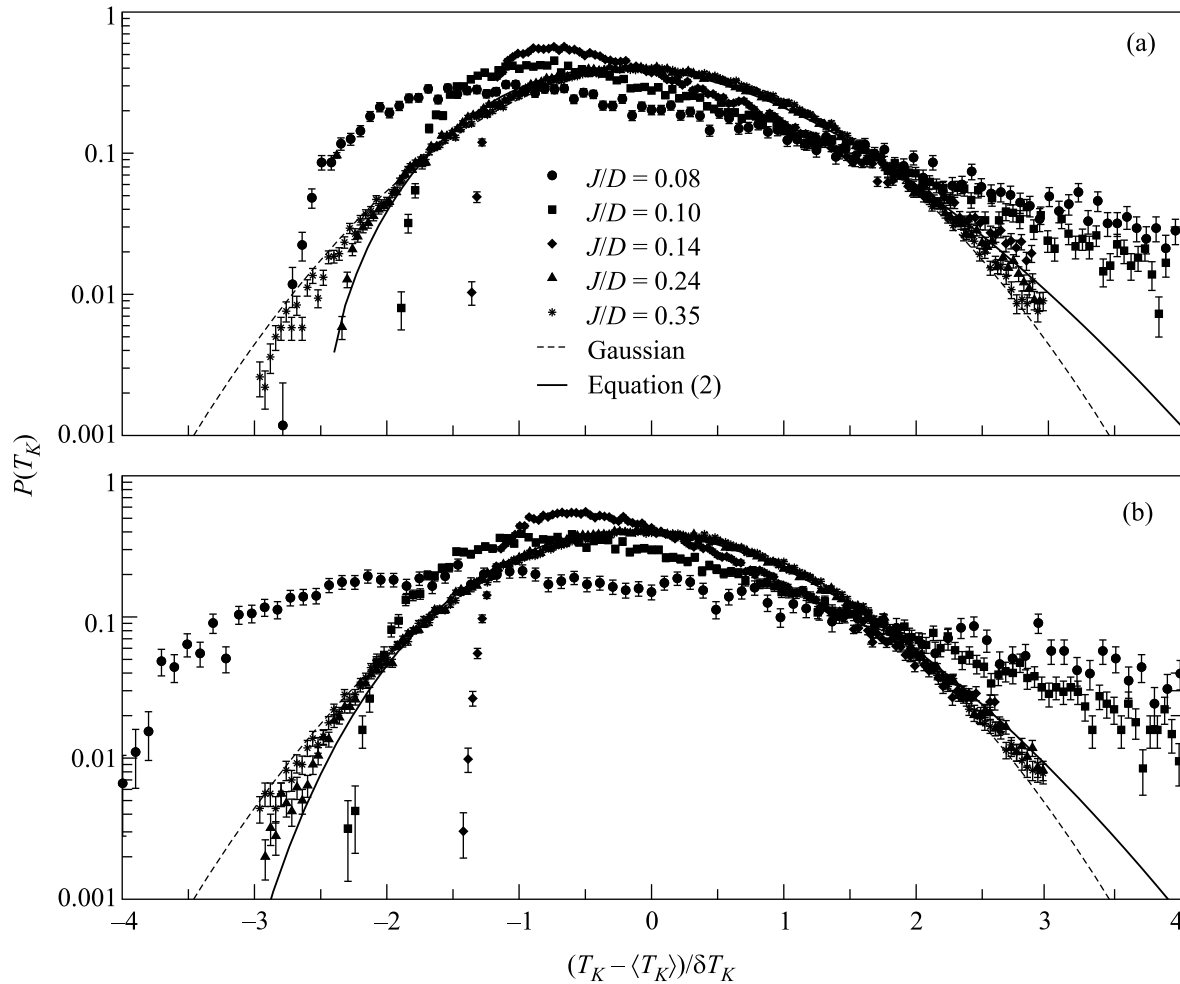


Fig.1. The distribution of  $T_K$  as scaled with its standard deviation  $\delta T_K$  for GOE (a) and GUE (b). Events corresponding to  $T_K = 0$  were neither included in the distributions nor in the average and the standard deviation used in the shifting and rescaling of  $T_K$ . The dashed line corresponds to a Gaussian with zero mean and unit variance. The solid line corresponds to Eq. (2) for  $J/D = 0.24$

where  $N$  is the number of states in the band of width  $D$ . Here, we have used the rescalings  $x = D/J$  and  $\kappa = T_K/\Delta$ , with  $x_n = L^d |\psi_n(\mathbf{r})|^2$  equals the probability density of the  $n$ -th eigenstate and  $s_n = (E_n - E_F)/\Delta$  is the eigenenergy relative to the Fermi energy in units of  $\Delta$ . The number of electrons in the Kondo box is taken to be even. Equation (1) is valid in the weak coupling regime, when  $T_K \ll D$ . It coincides with the self-consistent solution of the one-loop renormalization group (RG) equation [8, 10] up to the tanh factor, which accounts for the finite-temperature occupation. While the approximations involved in deriving Eq. (1) fail to describe the Kondo system below  $T_K$ , Eq. (1) does yield the correct low-energy scale in the effective Fermi liquid theory, where  $T_K$  determines the Landau parameters [11]. Nevertheless, for finite systems, there may be deviations from one-parameter scaling due to sample-to-sample mesoscopic fluctuations [12].

In order to gain insight on the statistical properties of  $T_K$  over a wide range of values of  $J$ , we solved Eq. (1) numerically and compared the resulting distribution and moments of  $T_K$  with approximate analytical expressions appropriate to the different regimes already mentioned.

*Random Matrix Theory.* For  $g \gg 1$ , the energy levels within an energy window smaller than  $E_c$  obey the Wigner-Dyson distribution of RMT [13]. The eigenenergies  $s_n$  and eigenfunction intensities  $x_n$  fluctuate independently. The latter are themselves uncorrelated and obey the Porter-Thomas distributions [13, 3]. Figure 1 shows the distribution of  $T_K$  obtained from the unfolded spectrum of random matrices of size  $N = 500$  using 500 realizations for each Gaussian ensemble, namely orthogonal (GOE) and unitary (GUE). The Fermi energy is taken to be in the middle of the band. The dependence of the average  $\langle T_K \rangle$  on  $J$  at  $\langle T_K \rangle > \Delta$  is found to coincide with the clean case result, which

is given by  $T_K^{(0)} \approx 0.57 D \exp(-D/J)$  for a spectrum of equally spaced levels. For small  $J$ ,  $T_K$  approaches  $\Delta$ ; in a system without fluctuations of energy levels and wavefunctions, it would turn abruptly to zero at  $J^* \approx D/[\ln(2N) + 0.58]$  in a nonanalytical fashion,  $T_K^{(0)}(J \rightarrow J^*) = -\Delta/\ln[(D/J^* - D/J)/4]/2$ . However, fluctuations are important at  $\langle T_K \rangle < \Delta$ , leading to non-zero values for  $T_K$  below the clean-limit threshold  $J^*$ . In Fig.1, the distributions of Kondo temperatures for the GOE and GUE are shown for different values of  $J$ . The Kondo temperatures are shifted and rescaled in order to facilitate the comparison with a Gaussian distribution. The latter might be expected from a naive use of the central limit theorem: Since wave function amplitudes corresponding to distinct eigenstates are uncorrelated in RMT,  $T_K$  is determined by a sum of independent random variables when energy level fluctuations are neglected. However, even when  $\langle T_K \rangle$  exceeds  $\Delta$  there are deviations from the Gaussian behavior. This is in agreement with approximate analytical results obtained by performing the average over the wave function amplitudes exactly but assuming equally spaced energy levels and  $T_K \gg \Delta$ , when we find in the limit of large number of levels  $N$  in a straight forward but lengthy derivation [14]

$$P(T_K) \sim \frac{1}{\Delta} \exp \left[ \beta \kappa \ln \left( \frac{e\kappa_0}{\kappa} \right) - \beta \kappa_0 \right], \quad (2)$$

where  $\kappa_0 = T_K^{(0)}/\Delta$  and  $\beta$  denotes the ensemble ( $\beta = 1$  for the GOE and 2 for the GUE). This distribution yields  $\langle T_K \rangle = T_K^{(0)}$ , independent of  $\beta$ . For  $T_K \approx T_K^{(0)}$ , it is approximately Gaussian with a width  $\delta T_K \beta = \sqrt{\Delta T_K^{(0)}/\beta}$ , in agreement with an earlier derivation [15], but note that the distribution is not log-normal as was incorrectly concluded, there. The ratio between the variance in the orthogonal and unitary regime is thus  $\sqrt{2}$  for  $T_K^{(0)} > \Delta$ , in agreement with numerical results [14]. When time-reversal symmetry is present, the energy level repulsion is weaker and the tendency to localization stronger, resulting in a wider distribution than in the unitary regime [16]. Within RMT, the width is found to vanish in the thermodynamic limit, when  $\Delta \rightarrow 0$  [15].

**Anderson model.** Going beyond RMT, stronger deviations from the Gaussian behavior can be expected due to correlations in the wave functions. In order to describe the conduction electrons, we use the noninteracting tight-binding Anderson model with nearest-neighbor hopping  $t$  and random site energies drawn independently from a box distribution of width  $W$  centered at zero. We assume each single-electron eigenstate to be spin degenerate and consider  $20 \times 20$  square lattices

with periodic boundary conditions. The Hamiltonian was diagonalized and the eigenenergies and eigenvectors were used to determine  $T_K$  through Eq. (1) without unfolding of energy levels. The disorder amplitude  $W$  and the parameter  $g$  are related in the Born approximation by  $1/g = \frac{\pi}{48} W^2/E_F t$ , where the lattice constant is  $a = 1$  ( $N = L^2$ ) and  $\Delta = 8t/N$ . This expression is valid for weak disorder  $g > 1$  and for a flat density of states. We chose  $E_F = 2t$  to avoid traces of the van Hove peak at the band middle. Only time-reversal symmetric cases were studied. The resulting distribution of  $T_K$  is shown in Fig.2, where a non-Gaussian

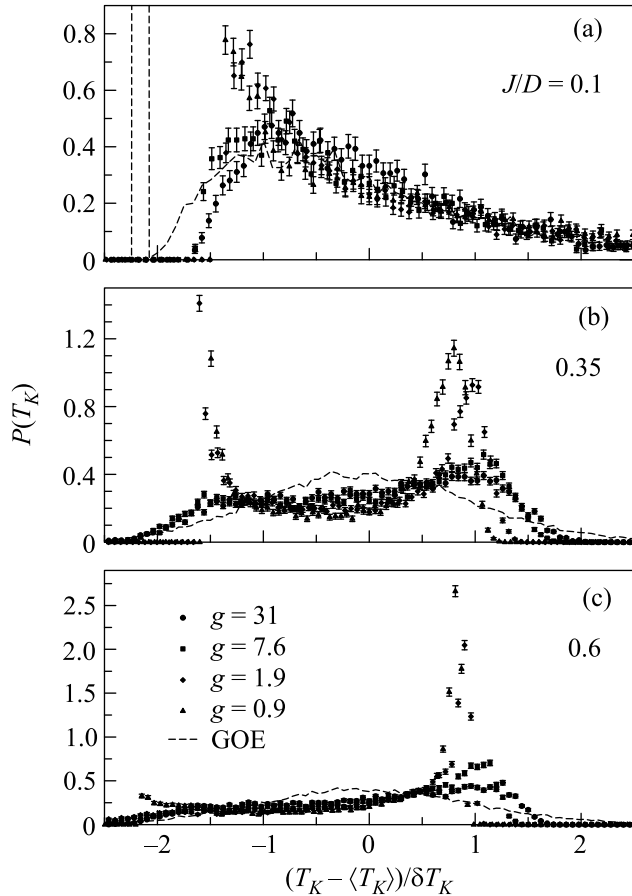


Fig.2. The distribution of  $T_K$  with eigenstates of a  $20 \times 20$  Anderson model without magnetic field (solid symbols) compared with the distribution for the GOE with  $N = 400$  (dashed line). For the Anderson model, 4,000 realizations were used for each value of  $g$

shape is evident. The GOE and Anderson model distributions only agree at weak disorder and for  $J \ll D$ , Fig.2a. For larger  $J$  (see Fig.2b), the distribution remains non-Gaussian for all disorder strengths, while the GOE distribution corresponding to the same value of  $J/D$  is almost Gaussian. The disordered Anderson

model presents a bimodal structure in  $P(T_K)$  that is absent in the RMT case. That structure is strongly pronounced in the strong disorder regimes, with two distinct peaks clearly visible. As  $J/D$  increases further, weight is transferred from the low- $T_K$  to the high- $T_K$  peak (see Fig. 2c).

Analytical information on the fluctuations of  $T_K$  can be obtained by relating it to the correlation function of the LDOS [3]. Defining  $T_K^{(0)}$  by Eq. (1) with average DOS  $\nu$ , we can rewrite Eq. (1) in terms of the LDOS as [17]

$$\ln \frac{T_K}{T_K^{(0)}} = \int_{-E_F}^{D-E_F} dE \frac{\delta\rho(E_F + E, \mathbf{r})}{2\nu E} \tanh\left(\frac{E}{2T_K}\right). \quad (3)$$

Expanding  $T_K$  to second order in  $\delta\rho = \rho - \nu$ , we can relate it to the correlation function of LDOS,  $R_2^{\text{LDOS}}(\omega) = \langle \rho(\mathbf{r}, E) \rho(\mathbf{r}, E + \omega) \rangle / \nu^2$ . While in RMT there are no correlations between wave functions at different energies, in the disordered Anderson model they are of order  $1/g$ ,

$$L^2 \langle |\psi_n(\mathbf{r})|^2 |\psi_m(\mathbf{r})|^2 \rangle \equiv f_{nm}(\omega) = 1 + \frac{2}{\beta} \text{Re} \Pi_\omega, \quad (4)$$

independently of the states  $n, m$  ( $n \neq m$ ). The correlation is stronger in the time-reversal symmetry case. The dependence on disorder is determined by a sum over diffusion modes, yielding, for  $L > l$  and in two-dimensions (2D),

$$\text{Re} \Pi_\omega = \frac{1}{4\pi g} \ln \left( \frac{1/4\tau^2 + \omega^2}{E_c^2 + \omega^2} \right). \quad (5)$$

In terms of the spectral correlation function,  $R_2(\omega) = \langle \rho(E) \rho(E + \omega) \rangle / \nu^2 - 1$ , the correlation function of LDOS reads

$$R_2^{\text{LDOS}}(\omega) = R_2(\omega) f_{nm}(\omega) + \delta(\omega/\Delta) f_{nn}(\omega). \quad (6)$$

For  $\omega < E_c$ ,  $R_2(\omega)$  has an oscillatory correction of order  $1/g^2$  to the leading RMT term [3]. For  $\omega > E_c$ , the oscillatory part of the spectral correlation function decays exponentially, while there is a correction of order  $1/g^2$  which decays like  $1/\omega$  without oscillations [3]. Expanding the exponent to second order in  $\delta\rho$ , we find

$$(\delta T_K)^2 = 2 \left( T_K^{(0)} \right)^2 \left[ S_\beta \left( \tau, T_K^{(0)} \right) + C_\beta \left( \tau, T_K^{(0)} \right) \right], \quad (7)$$

where  $S_\beta$  arises from the spectral self-correlation term proportional to  $\delta(\omega/\Delta)$  and is of order  $\Delta/T_K$ . To this order we also obtain  $\langle T_K \rangle = T_K^{(0)} + (\delta T_K)^2 / 2T_K^{(0)}$ . The

decaying parts of the spectral correlation function and the correlations of wave functions yield

$$C_\beta(\tau, T_K) = \int_{-E_F}^{D-E_F} \frac{dE dE'}{8 E E'} \tanh \frac{E}{2T_K} \tanh \frac{E'}{2T_K} \times \\ \times [-1 + R_2(E - E') f_{nm}(E - E')]. \quad (8)$$

In the thermodynamic limit  $E_c \rightarrow 0$ , the leading term in Eq. (8) arises due to the correlations between wave functions; to leading logarithmic order, we obtain

$$C_\beta(\tau, T_K) = \frac{1}{6\pi\beta g} \ln^3 \left( \frac{E_F}{gT_K} \right), \quad (9)$$

which is valid for  $D/J \gg \ln g$ . Thus, the width of the distribution is finite in the thermodynamic limit and the deviation of the average Kondo temperature from the clean limit is stronger than expected from Anderson's theorem when applied to the Kondo problem, in contrast to an earlier prediction [18]. A finite  $\delta T_K$  will have implications to the thermodynamic properties of disordered metals with magnetic impurities, as well as to heavy fermion materials. An earlier work [19] considered the effect of fluctuations of LDOS as well, but obtained an even larger effect by disregarding that wave function correlations are only of order  $1/g$ .

The expressions for  $\langle T_K \rangle$  and  $\delta T_K$  based on Eq. (7) are compared with results from the numerical simulations in Fig. 3. They are found to agree qualitatively with the dependence on the disorder strength  $g$  seen in the simulations for different values of  $J$ . It is only at  $g \gg 1$  and  $\langle T_K \rangle \gg \Delta$  that  $\langle T_K \rangle$  is insensitive to disorder. In general, for weak disorder, there is good agreement between GOE and Anderson model results for  $\delta T_K$  (not shown). At stronger disorder, fluctuations in the Kondo temperature increase in amplitude due to the increase of correlations in the LDOS at different energies over an interval of order of  $1/\tau$ , Eq. (4).

*Free moments.* The long tail toward small- $T_K$  indicates that disorder enhances the probability of having free magnetic moments at low temperature. This is confirmed in Fig. 4, where we show the probability of finding no solution to Eq. (1) as a function of  $J/D$ . We note that for  $T_K < \Delta$  only the two closest levels to the Fermi energy are strongly coupled to the magnetic impurity. In the clean limit, the probability of free moments is a sharp step function of  $J$ , namely,  $P_{\text{free}}(J \leq J^*) = 1$ , and  $P_{\text{free}}(J > J^*) = 0$ . When disorder is present,  $J^*$  fluctuates due to the random wave function intensities  $x_n$  and level spacings  $s_n$  of the two closest levels to the Fermi energy [14]. The probability that the level spacing at the Fermi energy is of the order of  $T_K^{(0)}(J)$  is exponentially small for  $J > J^*$ . However, in Fig. 4 we see that

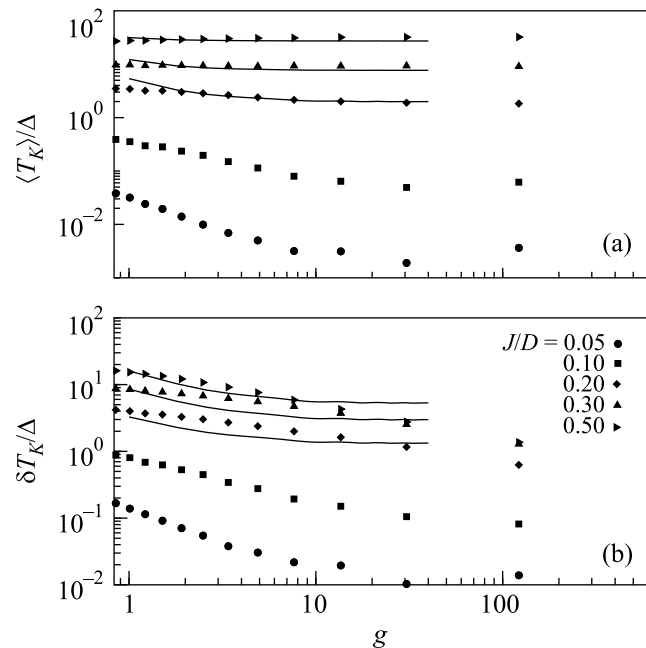


Fig.3. Dependence of average Kondo temperature  $\langle T_K \rangle$  (a) and standard deviation (b) in the Anderson model on the dimensionless conductance  $g$  for different values of  $J/D$ . Conditions are identical to Fig.2, with averages taken over 500 realizations. The solid lines correspond to Eq. (7) keeping  $E_c$  and  $\Delta$  finite. Notice that since we assume  $E_c\tau < 1$ , the lines are only presented for  $1 < g < 20$

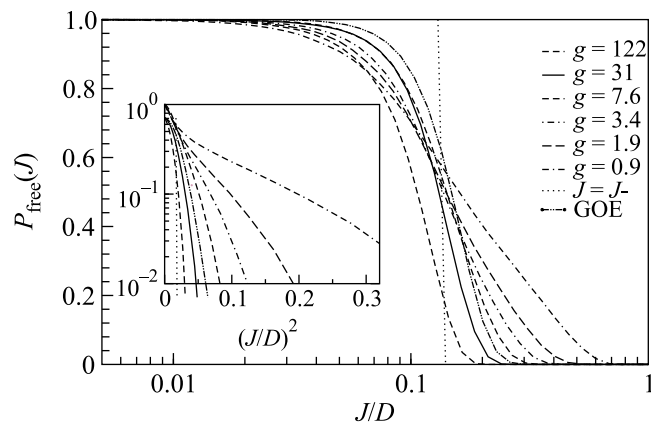


Fig.4. Probability of free moments as a function of  $J$  for the Anderson model at different disorder strengths  $g$  (same conditions of Fig.3). Also shown are the numerical result for the GOE with  $N = 400$ .  $J^*$  was estimated for a flat band

the decay with  $J$  is slower than that, indicating that the fluctuations of wave functions are crucial in the appreciable increase in the probability of free moments for  $J > J^*$ .

In the absence of an external magnetic field, magnetic impurities can break time-reversal symmetry since

the spin dynamics of the magnetic impurities is slow compared to the time scale of the conduction electrons [20]. For a mesoscopic sample at  $T > E_c$ , weak localization corrections are suppressed when  $X_s^{\text{WL}} = 1/E_c\tau_s$  exceeds 1 [20]. At finite temperature, the spin scattering rate  $1/\tau_s$  is renormalized by the Kondo correlations: It is small at both  $T \gg T_K$  and  $T \ll T_K$ , having a maximum at around  $T = T_K$  [21], which is given by a fraction  $\alpha$  of the unitary limit of the scattering cross section. In 2D, one finds  $1/\tau_{s\text{max}} = \alpha n_M v_F / 2k_F$ , where  $n_M$  is the concentration of magnetic impurities, with  $\alpha \approx 0.2$  [22, 23]. Thus, the maximum value of the crossover parameter is given by  $X_{sM} = \alpha N_M / g$ , where  $N_M = n_M L^2$  is the number of magnetic impurities in the sample. When there are only a few magnetic impurities,  $N_M < g$ ,  $X_s < 1$ , the metallic grain is in the orthogonal regime. Increasing the concentration of magnetic impurities increases the parameter  $X_s$ , thus decreasing the width of the distribution of Kondo temperatures. The maximum in the temperature dependence of the spin scattering rate results in a plateau of the dephasing time [21]. Mesoscopic Au wires with Fe impurities at concentrations  $n_S = 3 - 10$  ppm have a Kondo temperature  $T_K \approx 300$  mK [21]. While both  $E_c$  and  $\Delta$  are found to be of order  $10^{-7}$  K, and thus completely negligible in such wires, we can estimate that  $J/D = 0.074$  is much larger than  $J^*/D = 0.035$ . Therefore, we obtain from Eq. (9) that the width of the distribution of Kondo temperatures  $\delta T_K$  is of the same order as  $\langle T_K \rangle$ , with  $\langle T_K \rangle = 300$  mK.

To conclude, the distribution of Kondo temperatures has a finite width in disordered metals due the correlations of eigenfunctions within an energy interval of width  $1/\tau$ . Thus, the presence of free magnetic moments in disordered metals at temperatures below the average Kondo temperature is expected to yield a finite contribution to the electron dephasing rate and to the thermodynamic properties of disordered metals in the presence of localized magnetic moments [24].

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*Note:* After the submission of this work we learned that a result similar to Eq. (9) was recently obtained independently by Micklitz and collaborators [25]. After

the submission we also learned of the work by Cornaglia and collaborators who also studied numerically the distribution of the Kondo temperature, and also found a peak at small  $T_K$  [26].

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