Two-Impurity Kondo Physics

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In this essay we review the two-impurity Kondo problem. Our goal is to proceed in some pedagogical manner; hence, we start with a discussion of the (single-impurity) Kondo problem. We will discuss the fixed points of the model and motivate the Anderson model as a natural generalization of this system. Upon introducing a second magnetic impurity, we will discuss the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction. Finally, we will discuss recent experimental efforts made to explore the phase diagram of the two-impurity Kondo model.

1 The Kondo Problem

[1] Beginning in the 1930s, physicists became perplexed by the observed change in a metal's resistivity when doped with magnetic impurities — the resistivity of most metals is a monotonically increasing function of temperature; however, when diluted with magnetic atoms, the function becomes *convex* (Fig. 1).[1] In the 1960s, Jun Kondo was able to explain this phenomenon via perturbation theory;[2] however, the theory fails for temperatures below the so-called *Kondo temperature* T_K, because of logarithmic divergences in his resulting equation — the hunt that ensued to explain the low energy properties of the model is known as the *Kondo problem*.[1]

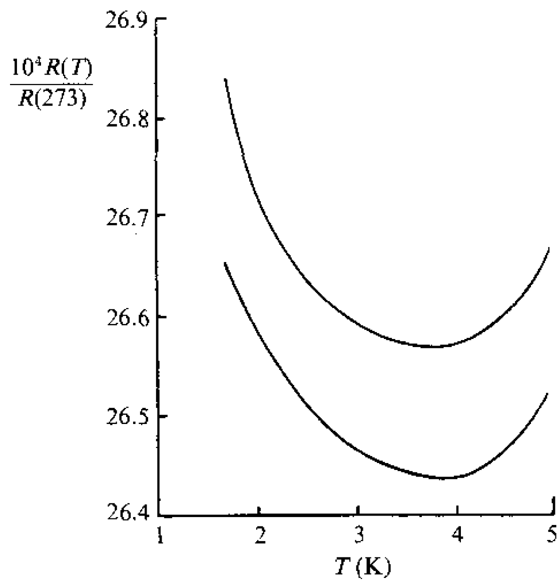

FIGURE 1: Reproduction of the first figure in [1]. This shows the actual resistivity minimum for a dilute magnetic alloy, in this case Au.

The model's Hamiltonian, known as the *Kondo Hamiltonian*, considers the coupling between the magnetic moments of a spin$-1/2$ magnetic atom (MA) and the conduction electrons in a metal:[2]

$$\mathcal{H} = \mathcal{H}_0 + J\,\mathbf{S} \cdot s(r_0) \tag{1.1}$$

where $\mathcal{H}_0$ is the Hamiltonian of the host metal, $\mathbf{S}$ is the spin operator of the MA (which is located at position r_0), $s(r)$ is the spin density operator of the host metal at position r, and J is a coupling constant.[3, 4] Specifically,

$$\mathcal{H}_0 = \sum_k \epsilon_k c^\dagger_{k,\sigma} c_{k,\sigma} \tag{1.2}$$

$$s_\mu(r) = \psi^\dagger_\sigma(r) \frac{\tau^\mu_{\sigma,\sigma'}}{2} \psi_{\sigma'}(r) \tag{1.3}$$

[1]This section is, for the most part, a very brief summary of the first few chapters of Hewson's book *The Kondo Problem to Heavy Fermions*.[1]

[2]Note: the notation we adopt borrows heavily from [1, 3–5].

where $c^{\dagger}_{k,\sigma}$ creates a spin$-\sigma(=\uparrow,\downarrow)$ conduction band electron with energy ϵ_k, $\psi^{\dagger}_{\sigma}(r)$ is an electron *field operator*[3] which creates a spin$-\sigma$ conduction band electron at position r, and τ^{μ} ($\mu = x, y, z$) are the Pauli matrices. [Note: we have employed the Einstein summation convention on the spin indices.]

Just by inspection of the Hamiltonian written in Eq. 1.1, one is able to see that the impurity term has some interesting implications. Notice that if $J > 0$, this terms suggests that it is energetically favourable for the spin of the MA to align *oppositely* to the spin of the nearest conduction band electron — the coupling is said to be antiferromagnetic (AFM); whereas, if $J < 0$, it suggests it is energetically favourable for the spins to align *parallel* — the coupling is said to be ferromagnetic (FM).[1]

FIGURE 2: Two regimes of the Kondo model. The energetics of the Hamiltonian suggests an AFM state for $J > 0$ and a FM one for $J < 0$. In this illustration, the sphere is meant to depict a MA and the solid line a conduction electron energy level. The red arrows are meant to be the spins associated with each site.

In the 1970s, slightly over a decade after Kondo's seminal work, Kenneth G. Wilson was able to access the low-temperature properties of the Kondo Hamiltonian in a non-perturbative way through his development of the *numerical renormalization group technique* (NRG).[7] The details on how one places the Hamiltonian on a lattice, performs the so-called *logarithmic discretization*, and carries out the *renormalization group* (RG) transformation for the Kondo problem can be found in [1, 3, 7].

Wilson was able to show that, in addition to the known *fixed point* at $J = 0$, there was also a fixed point in the model at $J = \infty$. Through his analysis of the problem, it became clear that the fixed point at $J = \infty$ was *stable* to RG transformations, whereas the fixed point at $J = 0$ was *unstable*.[1] Namely, the renormalization group flow is driven away from $J = 0$ and towards $J = \infty$:

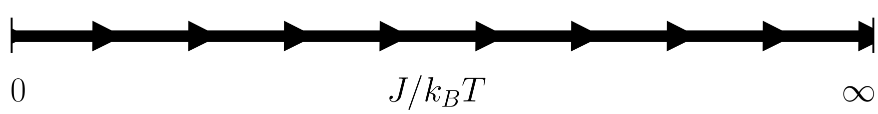

FIGURE 3: Illustration of the RG flow in the Kondo problem.

For $T \to 0$ or $J \to \infty$, the coupling becomes strongly antiferromagnetic. At $T = 0$ or $J = \infty$ the MA becomes locked into a spin-singlet state with the conduction cloud — the identity of the MA is lost as it is *screened* by the conduction electrons.[1]

[3]A nice explanation of the second quantization formalism is provided in chapter 1 of Fetter and Walecka's text *Quantum Theory of Many-Particle Systems*.[6]

Physically this means that at low energies one should observe an increase in the density of states on the MA near the Fermi energy of the metal — for this reason, the resulting characteristic signal is called the *Kondo resonance* (Fig. 4).[1, 8]

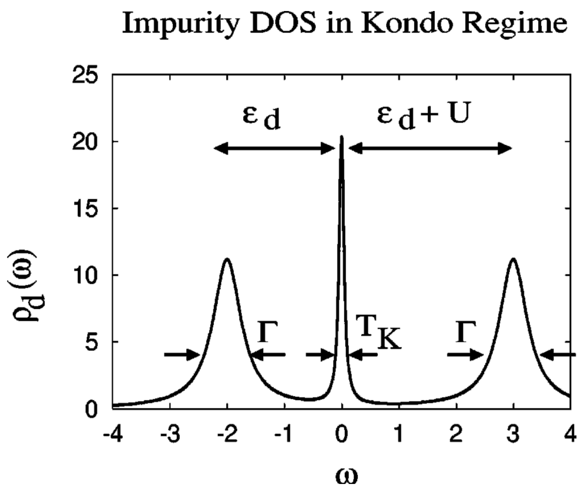

FIGURE 4: Illustration of the Kondo resonance; namely, the center peak (taken from [8]). Most parameters in the plot correspond to energy scales to be discussed in the next section; however, the Kondo temperature T_K corresponds to the central width in the image.

2 The Anderson Model

A rather transparent model for a spin$-1/2$ magnetic moment in a host metal is the *single-impurity Anderson Model* — here one considers an atomic d-orbital or f-orbital embedded inside a metal.[3, 4] A Hamiltonian of this form has the structure

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{MA}} + V \tag{2.1}$$

where $\mathcal{H}_0 (= \sum_k \epsilon_k c^\dagger_{k,\sigma} c_{k,\sigma})$ is the bare Hamiltonian of the metal (as discussed in the Kondo problem), $\mathcal{H}_{\text{MA}}$ is the MA's bare Hamiltonian, and V takes into account interactions between the atom and the metal. Specifically, consider

$$\mathcal{H}_{\text{MA}} = \epsilon_f f^\dagger_\sigma f_\sigma + U f^\dagger_\uparrow f_\uparrow f^\dagger_\downarrow f_\downarrow \tag{2.2}$$

$$V = T \left[f^\dagger_\sigma \psi_\sigma(r_0) + \psi^\dagger_\sigma(r_0) f_\sigma \right] \tag{2.3}$$

where in Eq. 2.2, $f^\dagger_\sigma$ and f_σ are fermionic creation and annihilation operators for electrons of spin$-\sigma(=\uparrow,\downarrow)$ in the atomic d-orbital (or f-orbital), ϵ_f represents the energy of an electron in this active orbital, and the on-site energy scale $U(> 0)$ is included to model the repulsive Coulomb interaction between electrons.[3, 4]

Eq. 2.3 captures one of the simplest events we can imagine taking place between two electrons — changing places! Namely, this term performs the task of destroying a conduction electron at the site of the MA (via $\psi(r_0)$) and then creating an electron on the MA's active orbital (via $f^\dagger_\sigma$) and vice versa — T represents the probability amplitude of this occurring.

There are a few different energy regimes associated with the Anderson model; these are the so-called *local moment*, *intermediate valence*, and *empty orbital* regimes.[1] In the empty regime, one has either $\epsilon_f \gg E_F$ or $\epsilon_f + U \ll E_F$ where E_F is the Fermi energy of the metal — in both cases the MA readily ionizes (losing an electron in the first case and gaining one in the second). In the intermediate valence regime, one has either $\epsilon_f \approx E_F$ or $\epsilon_f + U \approx E_F$. However, the most useful case for us is the local moment regime in which $\epsilon_f \ll E_F \ll \epsilon_f + U$ — in this case, notice that it is energetically *costly* for the MA to ionize; namely, it costs energy for the electron on the MA orbital to enter into the Fermi sea, but it is also costs energy for a conduction electron to join the MA. Hence, one can see why it is possible for a *localized* magnetic moment to survive in this environment.[1]

Not that long after Kondo completed his calculation of the system's resistivity, Schrieffer and Wolff showed that in the local moment case ($\epsilon_f \ll E_F \ll \epsilon_f + U$), the Kondo model follows from the Anderson model.[9] Their result was that the coupling constant in the Kondo Hamiltonian is a function of the the energy scales in the Anderson model and is proportional to

$$J \propto \frac{U}{\epsilon_f(\epsilon_f + U)} \tag{2.4}$$

3 The Two-Impurity Kondo Problem

Let us now consider a logical extension to the Kondo problem: let us place a second MA in the metal — this is the so-called *two-impurity Kondo problem.*[5] To model such a system, an appropriate starting point is to consider the Anderson model we just discussed, but with additional terms for the extra spin$-1/2$ magnetic moment:[3, 4]

$$\mathcal{H} = \mathcal{H}_0 + \sum_{l=1}^{2} [\mathcal{H}_{\mathrm{MA},l} + V_l] \tag{3.1}$$

where $\mathcal{H}_0$ is the same as before and, as the notation implies, $\mathcal{H}_{\mathrm{MA},l}$ is the Hamiltonian of MA l with V_l representing interactions between this impurity and the host metal. If both of the atoms are of the same variety we can write the terms explicitly as

$$\mathcal{H}_{\mathrm{MA},l} = \epsilon_f f^\dagger_{l\sigma} f_{l\sigma} + U f^\dagger_{l\uparrow} f_{l\uparrow} f^\dagger_{l\downarrow} f_{l\downarrow} \tag{3.2}$$

$$V_l = T \left[f^\dagger_{l\sigma} \psi_\sigma(r_l) + \psi^\dagger_\sigma(r_l) f_{l\sigma} \right] \tag{3.3}$$

where ϵ_f, U, T, and $\psi_\sigma(r)$ are the same as before — we have simply added labels to the fermionic operators $f_{l\sigma}$ to distinguish which of the atoms (located at positions r_1 and r_2) they belong to. A natural Kondo model for the two-impurity system is obtained in the "Schrieffer-Wolff limit" of this Hamiltonian:[3, 4]

$$\mathcal{H} = \mathcal{H}_0 + J_K \, \mathbf{S}_1 \cdot s(r_1) + J_K \, \mathbf{S}_2 \cdot s(r_2) \tag{3.4}$$

where J_K is a Kondo coupling constant, $\mathbf{S}_l$ is the spin operator of MA l, and $s(r_l)$ is the metal's spin-density operator at r_l.[4]

[4] Note: for simplicity, I have ignored the *direct exchange* interaction term discussed in [3], because the effective Hamiltonian we arrive at is of the same form; however, it should be included in general.

4 The RKKY Interaction

Based on the form of Eq. 3.4 one might expect that the low energy physics of the model corresponds to two MAs individually locked in a singlet state with the conduction electrons; however, we'll see the actual situation is far more complex. Upon integrating-out short wavelength conduction electron states, Jayaprakash et al. found that the effective Hamiltonian for the system is

$$\mathcal{H} = \mathcal{H}_0 + J\,\mathbf{S}_1 \cdot s(r_1) + J\,\mathbf{S}_2 \cdot s(r_2) + I\,\mathbf{S}_1 \cdot \mathbf{S}_2 \tag{4.1}$$

where, as a result of tracing out the higher energy degrees of freedom, one obtains updated coupling constants J and also introduces a new interaction between the atoms' spin operators — $\mathbf{S}_1 \cdot \mathbf{S}_2$ with a coupling constant I.[5] This coupling constant corresponds the so-called *Ruderman-Kittel-Kasuya-Yosida* (RKKY) interaction.[5]

This *indirect exchange* interaction is represented pictorially below in Fig. 5. As described in [10] (in the context of nuclear moments in a metal), the process can be thought of as follows: a conduction electron in the metal is able to interact with an electron occupying the first MA through their mutual Kondo interaction. The electron is scattered and this information can propagate, via the spin density of the metal, to the second MA where the same kind of interaction occurs. Hence, if both MAs exchange their spins with the conduction electrons, the net result of this process is that the two impurities have exchanged their spins with one another.

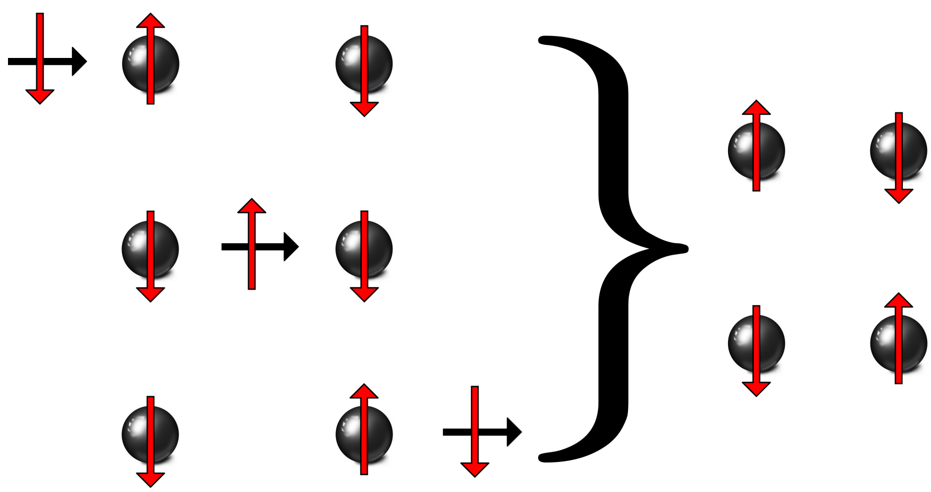

FIGURE 5: A cartoon picture showing how two magnetic moments, separated in space, can (indirectly) interact with one another through the conduction electrons in the host metal.

The RKKY interaction describes the coupling between two spin operators — so, as in the Kondo problem, there is an AFM ($I > 0$) and a FM ($I < 0$) regime associated with the model. However, there is also new physics here, because the RKKY interaction is dependent on distance; namely, by appropriately varying the separation distance between the two impurities, one can modify the interaction from an AFM one to a FM one and vice versa.[5]

This effect is depicted in Fig. 6 — one can see that when the two impurities are near one another, the interaction favours a FM state.[5] As discussed in [5], this makes sense physically, because as the two MAs get closer together, they start to share more and more of the same electrons; hence, in the limit that they are essentially touching, they will both be interacting with the same electron and thus have the same spin. As one of the impurities is moved further away, the effect diminishes. We see that for distances slightly over twice the inverse Fermi wavevector, the interaction switches sign and becomes AFM.

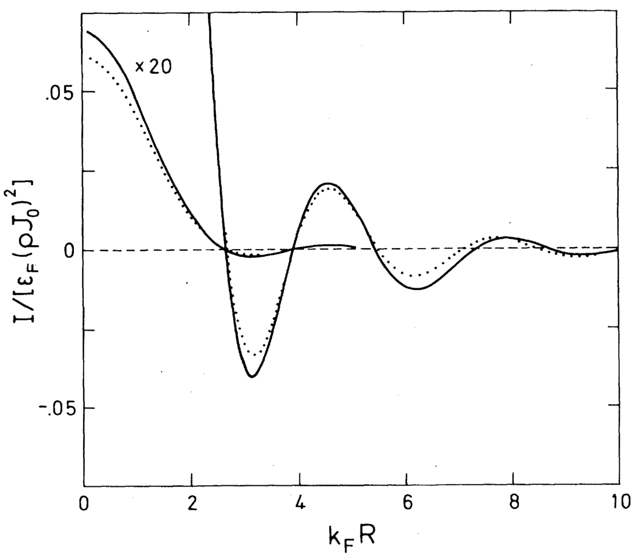

FIGURE 6: A plot showing the oscillatory nature of the RKKY interaction as the distance between MAs changes. One can see that, depending on their atomic separation, it is possible to achieve either a AFM or FM coupling. The image comes directly from [5].

5 Phases of The Two-Impurity Kondo Model

Suppose we have two MAs in a metal spatially separated such that $I > 0$ — i.e. they experience an AFM RKKY interaction. At low temperatures, we know the atoms also participate in the (AFM) Kondo screening effect with the electron sea. Thus, there is a competition between the two effects — will the MAs form a singlet with each other, or instead be effectively screened by the nearby sea of conduction electrons?

The two important energy scales in the problem are the Kondo temperature T_K, below which MAs are screened, and the interimpurity RKKY interaction strength I. We can attempt to characterize the phase of the system using the ratio I/T_K.[3] In the limit $I/T_K \to \infty$ the Kondo screening vanishes and the MAs enter into a spin-singlet

[5] There is no agreed upon sign convention for the coupling constants in the Kondo models, so naturally different authors use different signs (for example, [1] versus [5]). Compared to our notation, Jayaprakash et al. use the opposite sign for their coupling constants; hence, in our notation, Fig. 6 corresponds to $I < 0$ at short separation distances — i.e. the coupling is FM.

state with one another. Going the opposite direction and taking $I/T_K \to -\infty$ once again yields no Kondo effect, but now the MAs are in a spin-triplet state. Finally, consider taking the limit as $I/T_K \to 0$ — in this case the Kondo screening will be so strong (and the RKKY interaction so weak) that each atom will form a singlet state with the conduction electrons surrounding it.[3] A complete theory of the phase diagram of this model remains incomplete however.[3, 4]

Recently, Spinelli et al. were able to experimentally explore the phase diagram of the two-impurity Kondo model by using a *scanning tunnelling microscope* to perform differential conductance measurements of (magnetic) Co atoms in various configurations on a Cu_2N/Cu(100) substrate.[11] Their phase diagram is shown in Fig. 7. We see that when I/T_K is sufficiently positive or negative one does indeed find that the two impurities couple together in a spin-singlet or a spin-triplet state respectively (i.e. no Kondo effect is observed). Then, for smaller I/T_K ratios, Kondo screening becomes the operative physics.[11]

However, unlike what we have discussed so far, the group also placed the system in a magnetic field. In doing so, they were allowed to access a broader range of phenomena. As discussed in detail in their paper, they found that by only adjusting the external field they could take MAs with an AFM RKKY coupling (i.e. the system in the *two-impurity singlet* phase where the RKKY interaction is dominant) and make them behave as individual local moments (i.e. put the system in the *single impurity local moment* phase).[11] They were also able to show that an intermediate phase exists where the MAs are simultaneously in a spin-singlet state with one another while also being screened by the conduction electrons — namely, there exists a *two-impurity Kondo screening* phase.[11]

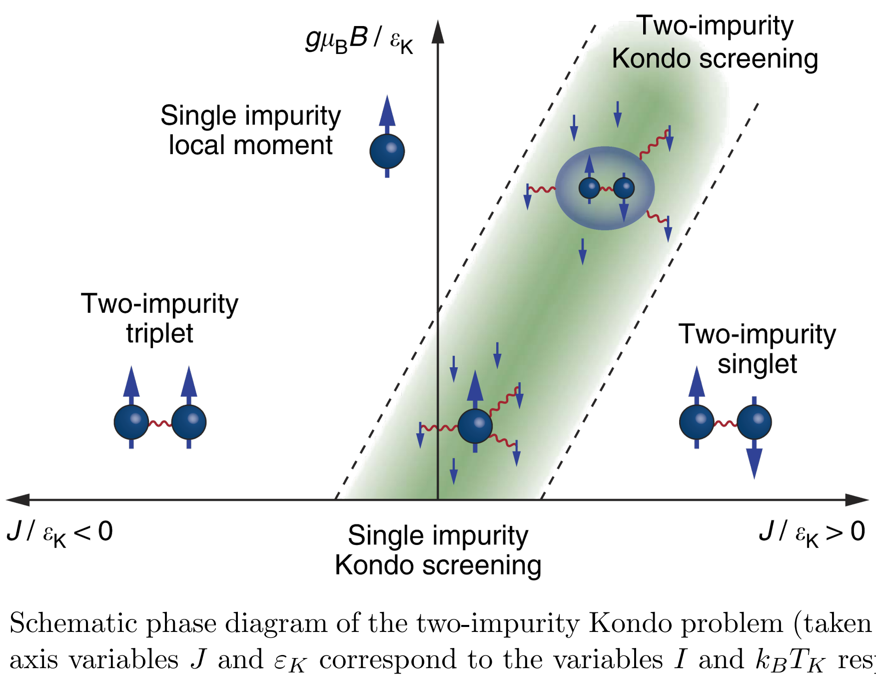

FIGURE 7: Schematic phase diagram of the two-impurity Kondo problem (taken from [11]). Here, the axis variables J and ε_K correspond to the variables I and $k_B T_K$ respectively.

6 Scanning Tunnelling Microscopy

Spinelli et al. conducted their experiments using the scanning tunnelling microscope (STM). The STM is a high resolution microscope that is designed to exploit the fact that tunneling currents between the so-called STM *tip* and atoms on the surface of a material are very sensitive to spatial variations.[8] The physical structure of the device is illustrated in Fig. 8.

In order to determine the differential conductance on the surface of a material, the device can perform a "spectroscopic" measurement;[6] It is important to note that the differential conductance is proportional to the electronic *local density of states*:

$$\frac{dI}{dV}(r, \epsilon) \propto \mathrm{LDOS}(r, \epsilon) \tag{6.1}$$

at energy ϵ with r being the position on the surface below the STM tip.[8]

It is because of this relation that the STM can detect Kondo screening; as we mentioned in the first section, a physical observable of the Kondo problem is the Kondo resonance in the density of states. An STM is able to detect if an impurity possesses this resonance or not, and hence this is "the main spectroscopic signature of Kondo atoms."[8]

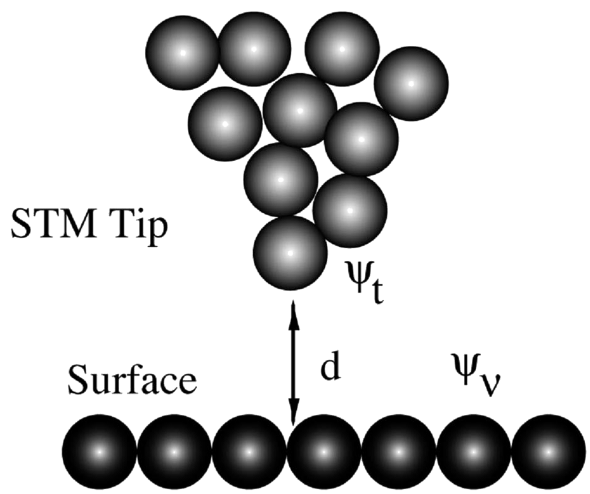

FIGURE 8: Visualization of an STM (taken from [8]). The STM tip (which is an atom in diameter) is brought within a few angstroms of a material's surface. By sweeping either the voltage or the position of the tip one can extract useful information about the material. By moving the tip closer still one is also able to move individual atoms around on the surface.

The STM is a remarkable device for another reason — it allows one to build nanoscale structures by translating individual atoms along the surface of a substrate. Using this method, it becomes possible to *manufacture* the desired RKKY interaction between two MAs.[11]

[6]The details of this operation are discussed in [8]. The device can perform other types of measurements too (also discussed in [8]), but as we are only interested in the differential conductance results in [11], we need not concern ourselves with those details.

Here we provide just a few more notes on the details of the experiment in [11]. To explore the physics of this problem, one needs their system to be well below the Kondo temperature. For the Co atoms used in this experiment, the Kondo temperature is given by $T_K = 2.6K$. Thus the experiments were carried out at a sufficiently lower temperature of $0.33K$.

Also, it is noted in the paper that they were unable to take their most AFM RKKY pair of Co atoms through the phase diagram by adjusting the magnetic field. The reason being that it would require a field strength larger than $9T$ (which their experimental setup was not equipped to handle).[11]

7 Discussion

In summary, we have discussed some of the many facets of the Kondo problem with a focus on its extension to two magnetic impurities. In doing this, we were able to briefly touch on some of the rich history of the problem, as well as the multitude of theoretical techniques which have been developed, in part, to handle aspects it. From the discovery of the resistance minimum in the 1930s to Wilson's groundbreaking work on the renormalization group in the 1970s and beyond, the Kondo problem and its extensions are intertwined in many areas of physics.

In sections 1 and 2, we introduced Jun Kondo's Hamiltonian for a spin$-1/2$ magnetic moment in a host metal and discussed its RG treatment. This lead us to the Anderson model of a magnetic moment — we saw the Kondo model was a subset of the Anderson model. Next, in sections 3 and 4 we added a second magnetic moment to the Anderson model and subsequently saw what happens when one integrates-out the short wavelength degrees of freedom; namely, the generation of a RKKY interaction between impurities. In section 5, we alluded to the outstanding problem in heavy fermion physics to understand the phase diagram of the two-impurity Kondo model.[3, 4] We then discussed, in a bit of detail, recent experimental investigations into the model's phase diagram and the discovery of a two-impurity Kondo screening phase in the presence of a magnetic field.[11] We concluded our discussion in section 6 by commenting a bit more on the tools used to experimentally probe this system.

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