# Observations of Kondo Scattering without Magnetic Impurities: A Point Contact Study of Two-Level Tunneling Systems in Metals

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We study singularities in the differential resistance of metal point contacts containing structural disorder. We find zero-bias features which are well described by theories in which two-level tunneling systems couple strongly to the conduction electrons to produce a Kondo resonance whose origin is not magnetic impurities. Away from zero bias, we observe abrupt resistance steps which indicate transitions in the multichannel Kondo condensate. We suggest that two-level systems are the explanation for many of the zero-bias anomalies seen in tunnel junctions and point contacts.

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The concept of the two-level tunneling system (TLS) provides the framework within which thermal and elastic properties of glassy materials are currently understood [1]. In recent years, theoretical efforts to describe the interaction between TLSs and conduction electrons in metals have produced intriguing results. Vladar and Zawadowski [2] showed that at low temperature $T$, the electron-TLS system may scale into a strong-coupling regime that is formally analogous to the Kondo condensate [3,4] for a magnetic impurity in a metal. Shortly thereafter, it was shown that if spin degeneracy is taken into account, the TLS-electron system should properly be described by a two-channel Kondo model [5]. Ideas involving TLSs and Kondo models have been put forth to explain properties of amorphous superconductors [6] and some heavy-fermion materials [7]. However, the basic results of the strong-coupling theories have never faced a direct test.

In this Letter we report measurements on metal constrictions so small that we are capable of probing electron scattering from individual defects by means of electrical resistance measurements. We are thus able to measure directly the properties of just a few TLSs within a metal. For a small applied voltage $V$, we measure logarithmic $T$ and $V$ dependences in the conductance, well described by strong-coupling Kondo models of TLSs. These conductance features do not have the magnetic-field dependence characteristic of the Kondo effect due to magnetic impurities. At larger $V$ we can probe the properties of TLSs interacting with a nonequilibrium electron distribution, a regime not yet considered by theory. As we sweep bias, we observe very sharp steps in the dc conductance, indicative of transitions in the electronic ground state within the sample.

The process by which we fabricate and characterize our constrictions has been described elsewhere [8]. We use electron beam lithography and reactive ion etching to form, in a silicon nitride membrane, a tapered hole whose minimum diameter may range from 3–15 nm. In ultrahigh vacuum ($<2\times10^{-10}$ torr) and at room temperature, we then rotate the membrane to expose both sides while evaporating metal to fill the hole and form the metal constriction. The effects described in this Letter have been seen in copper, aluminum, silver, and platinum constrictions [9]. All the data we present come from a single Cu constriction, allowing us to compare directly different aspects of a single device, but similar phenomena have been seen in tens of other samples.

Point contacts made by pressing together macroscopic pieces of metal have been studied for more than a decade. Both these devices and our constrictions are of interest because the electron mean free path can be much longer than the constriction diameter (in our copper films the mean free path is 180 nm at 4.2 K). As a result, when a voltage is applied across the constriction, a strongly nonequilibrium electron distribution is produced. Energy-dependent scattering mechanisms lead to a $V$-dependent resistance signal, providing a means for spectroscopic studies of scattering [10,11].

The signals which are the subject of this Letter occur only in constrictions which are cooled within hours of their evaporation, and the signals disappear if the samples are subsequently allowed to anneal at room temperature [8]. A low-$V$ differential conductance curve for an unannealed copper device is shown in Fig. 1. This signal falls in the class of zero-bias anomalies that have long been seen in point contacts [10] and tunnel devices [12], but have never been satisfactorily explained. With no applied magnetic field, the signal is similar to Kondo scattering from magnetic impurities [13]. However, the magnetic-field dependence demonstrates that the source of the scattering is not magnetic impurities. In a magnetic field $B$, the Zeeman splitting of the energy levels of a magnetic impurity suppresses Kondo scattering for $V<g\mu_B B/e$, where $g$ is the Landé $g$ factor and $\mu_B$ is the Bohr magneton. The result is a notch, a local peak, centered at $V=0$ in the conductance versus $V$ trace. This Zeeman splitting should be seen as long as the applied field is large enough that $g\mu_B B$ is larger than $k_B T_K$, where $T_K$ is the Kondo temperature [3]. For this case $g\mu_B B$ is in the regime where the energy dependence of the scattering is approximately logarithmic, which is true for our samples since

 

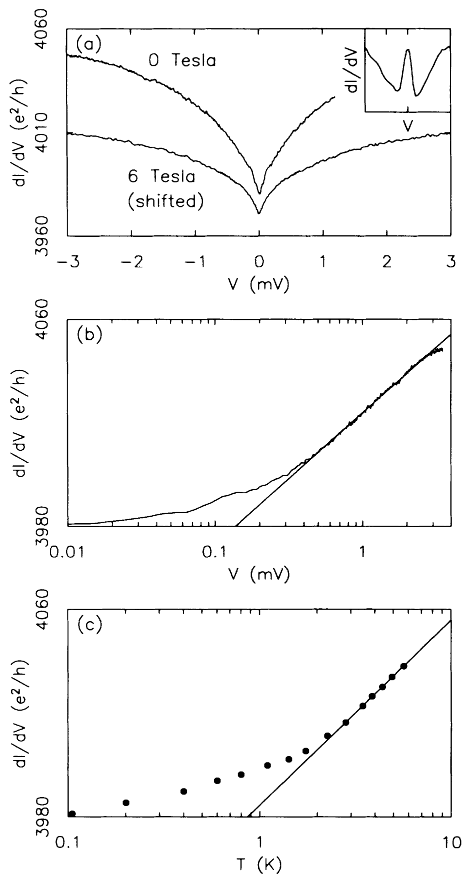


FIG. 1. (a) Differential conductance of a Cu constriction at $T=100$ mK, measured using ac modulation 0.02 mV rms superimposed on a dc voltage. The 6-T curve is shifted down by $20e^2/h$ for clarity. (b) $V$ dependence of the differential conductance for $B=0$ and $T=100$ mK. (c) $T$ dependence of the conductance for $B=0$ and $V=0$. Straight lines illustrate regions of logarithmic $V$ and $T$ dependences. Inset to (a): Conductance of Cu constriction with 200 ppm Mn at 100 mK, 4 T, showing suppression of magnetic-impurity Kondo scattering by a magnetic field. The conductance scale extends from $1025.4e^2/h$ to $1025.7e^2/h$, and the voltage scale from $-3$ to 3 mV.

for $g=2$ and $B=6$ T, $g\mu_B B=0.7$ meV. We observe no sign of a Zeeman splitting for these samples, although the splitting is clearly evident in samples intentionally doped with magnetic impurities [inset to Fig. 1(a)].

Recent theories demonstrate, however, that one does not need magnetic impurities in order to produce Kondo scattering. Theories of strong coupling between conduction electrons and a TLS also predict a logarithmic $T$ dependence in the conductance above a Kondo temperature, estimated to be as high as 5 K, with a low-$T$ roll-off to a limit produced by a unitarity limit of scattering [2]. We have observed zero-bias conduction dips which grow large enough to become observable at $T$ ranging from 10 K to 100 mK. The largest conductance features, including the one pictured in Fig. 1, show a logarithmic $T$ dependence above 2 K, and a roll-off to a slower decrease below this value. We note that the logarithmic $T$ and $V$ dependences that we observe extend not quite a full decade, a small enough range that other functional forms are not ruled out. Power-law fits to the data over the same ranges as the logarithmic fits require an exponent of less than 0 and greater than $-0.5$ for the $T$ dependence and between 0 and $-0.3$ for the $V$ dependence. The data well below the roll-off at 2 K may be fitted equally well by either a slower logarithm or the $1-aT^{0.5}$ form predicted by two-channel Kondo models [14]. However, our largest signals are likely due to several TLSs having a range of Kondo temperatures, so we do not expect to see a clear signature of the low-$T$ saturation in these data.

The magnitude of the conductance signal predicted for scattering from one TLS within a single-channel Kondo model [2] is $\Delta G \sim 2e^2/h$; but this model assumes that only two partial waves are dominant in the scattering. More complex multichannel models might well give an estimate several times larger. In order to explain the size of our largest signal, $\sim 70e^2/h$, we expect that we have on the order of 10 TLSs in the constriction region coupled strongly to conduction electrons and contributing to the resistance. We can estimate the diameter of the 6.4-$\Omega$ constriction based on the Sharvin formula [8] to be 13 nm, and there are $10^5$ Cu atoms within a sphere of this diameter about the constriction. The strongly coupled TLS density we estimate is therefore roughly of order $10^{-4}$/atom, about the same as estimates for the total density of TLSs in glassy systems [15]. Transmission electron microscopy studies of silicon constrictions with a geometry similar to ours indicate that dislocation networks may form in the constriction region during fabrication [16]. Dislocation jogs or kinks acting as TLSs are thus a possible cause of the signals we report.

Further evidence to support the idea that our zero-bias anomalies are due to TLSs comes both from studies of the annealing of the constrictions and from noise studies. The fact that the signals disappear on a time scale of days if a sample sits at room temperature (inset to Fig. 2) indicates that the signals are due to structural disorder which may anneal away. However, we know of no mechanism by which static disorder can give a $V$-dependent conductance in the 1-mV range, especially not such a large effect. This suggests a dynamic process due to TLSs. Also, time-dependent resistance studies performed previously on clean, well-annealed constrictions reveal directly the existence of slow two-level fluctuations caused by atomic motion [8,17]. The dynamics of those slow fluctuators are governed by thermally activated processes rather than tunneling, due to high potential barriers for defect movement, but, like tunneling systems, these fluctuators consist of atoms or groups of atoms moving between two metastable configurations. We suggest that before annealing each sample contains several fast (low potential barrier) tunneling states which may produce Kondo scattering, but that annealing tends to leave the sample with only a few two-level systems having high potential barriers so that only slow thermally activated transitions are common.

In macroscopic metallic glass samples, even if they contained a very large density of TLSs coupled strongly to electrons, on the order of $10^{-4}$/atom, a $T$-dependent resistance signal due to Kondo scattering from TLSs [2] would be masked by the larger signal from the $T$ dependence of weak localization and electron-electron interactions [18]. However, neither of these effects is significant in our samples. Both weak localization and interaction effects would give conductance changes of order $1e^2/h$, while the effects we see are much larger. Both effects also predict a positive low-field magnetoresistance in Cu, which has relatively strong spin-orbit scattering, while we observe a negative magnetoresistance. Furthermore, the absence of any visible universal conductance fluctuation [19] effects indicates that the electron mean free path in our samples is long enough that they are not in the diffusive regime where localization and interaction effects become important.

A dramatic and unanticipated effect occurs in most of our unannealed samples away from zero bias. Spikes occur in the differential conductance as shown in Fig. 2. These correspond to sudden steps in the dc conductance as a function of $V$. The true dc conductance change is less than $5e^2/h$, consistent with changes of electron scattering cross section on an atomic scale. The spikes appear in $V$-symmetric pairs and occur only in samples which also show a zero-bias dip in the conductance. These transitions are further associated with the zero-bias features by being suppressed in the same range of $T$, having magnetic-field dependence in the same range of field, and disappearing after room-temperature anneals. A minority, roughly 20%, of samples with zero-bias peaks do not show any observable $V \neq 0$ spikes. Usually more than one set of transitions occurs in a sample, and transitions have been observed at dc biases from 0.05 to 50 mV at zero magnetic field.

The $T$ dependence of these conductance transitions is quite striking. The voltage width of the transition in Fig. 2 decreases by a factor of 38 as $T$ is lowered from 7 to 4.2 K. At 4.2 K the full width at half maximum of the spike is only 0.03 mV, or only $\frac{1}{10}$ of $k_B T$. At lower $T$, the transition becomes abrupt and hysteretic. The size of the dc conductance jump is $T$ dependent, increasing in the example of the feature in Fig. 2 from $2.4e^2/h$ at 4.2 K to $4.8e^2/h$ at 2 K. Neither the $T$ dependence of the transitions nor the hysteresis can be explained by simple models of atomic rearrangements driven by $V$ [17]. A collective many-body mechanism involving positive feedback appears necessary. The magnetic-field dependence, discussed below, argues that almost certainly some sort of change in the ground state of the electrons is involved, associated with changes in the Kondo screening around the TLS. An analogous positive feedback behavior is seen in the suppression of the gap in superconductors by the creation of a nonequilibrium quasiparticle distribution [20].

The lower curve in Fig. 2 shows a typical behavior of the conductance transitions in a magnetic-field $B$ at high enough $T$ that there is no hysteresis. The transition bifurcates as a function of $B$, with the $B$ dependence of the voltage positions of the transitions shown in Fig. 3. After the bifurcation, as $B$ increases, both transitions move toward $V=0$ and broaden. The outer transition moves to $V=0$ with a quadratic dependence on $B$, while the inner transition moves in more quickly, with approximately a linear dependence. We suggest that this behavior may be a consequence of a two-channel Kondo Hamiltonian in the presence of a nonequilibrium electron distribution. In the two-channel Kondo picture of TLS behavior, an ap-

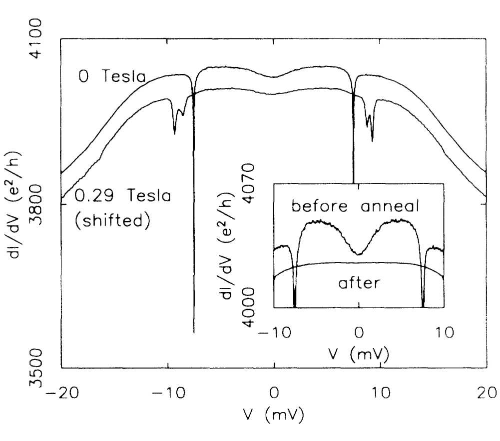


FIG. 2. Differential conductance of the Cu constriction at $T=4.2$ K showing sharp transitions in the conductance which bifurcate in a field of 0.29 T. The 0.29-T curve is shifted down by $40e^2/h$. The decrease in conductance beyond 10 mV is due to phonon emission. Inset: Magnified view of the same device for $B=0$ and $T=4.2$ K, before and after the sample annealed at room temperature for 1 week.

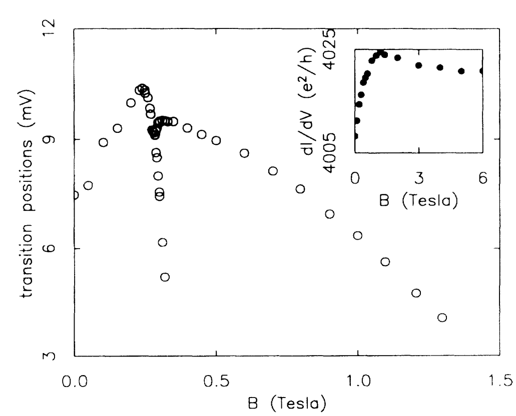


FIG. 3. Voltage positions of the conductance transitions at $T=4.2$ K as a function of magnetic field. Inset: The differential conductance for $V=0$ shows magnetic-field dependence in the same field range as the conductance transitions.

plied magnetic field breaks the time-reversal symmetry between electron-spin orientations [21], which could allow a single transition to split into two.

In summary, we have observed zero-bias dips, logarithmic in both $T$ and $V$, in the differential conductance of metal constrictions containing structural disorder. We explain these features as due to Kondo scattering from two-level tunneling states in the devices. We suggest that many of the logarithmic zero-bias peaks seen in point contacts and tunnel junctions which cannot be explained by magnetic impurities are due to this TLS-Kondo effect. At slightly higher biases we observe sharp features in the differential conductance apparently due to transitions in electronic screening around the TLSs. These transitions have a rich but not fully explained magnetic-field and $T$ dependence.

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