# Experimental Observation of Scattering of Tunneling Electrons by a Single Magnetic Moment

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(Received 20 September 1991)

Tunnel junctions formed between tungsten wires display very large zero-bias conductance peaks characteristic of Kondo-type magnetic scattering of tunneling electrons. No electron correlation effects are seen despite the fact that the system appears to be below the Kondo temperature. Time-dependent changes in the tunnel conductance are identified with the scattering behavior of a single local magnetic moment. This behavior can be switched reversibly by adjusting the stress in the junction.

PACS numbers: 73.40.Gk, 72.15.Qm, 75.30.Et

Zero-bias conductance peaks have long been seen in tunnel junctions. They were first observed in silicon *p-n* junctions [1] and shortly thereafter in metal-insulator-metal (MIM) junctions [2] and are believed to be caused by exchange scattering of the tunneling electrons by local magnetic moments within the tunnel barrier. Appelbaum [3] calculated the contribution to the tunnel current resulting from both spin-flip and Kondo-type scattering. The latter process is a third-order perturbation effect and is well known in dilute alloys of rare earths in noble metals. In macroscopic MIM junctions the zero-bias peak has been systematically studied by intentionally doping the junction with magnetic ions [4]. The maximum change in zero-bias conductivity is of order 10% and is obtained with about half a monolayer of magnetic ions. At higher coverages the interaction between the ions actually reduces the peak size.

In conventional planar junctions the magnitude of the effect is limited by the strength of coupling that can be obtained between the local moment and the conduction electrons in the electrodes. However, in "asperity" (see below) junctions characteristic of tunneling microscopes or the crossed-wire apparatus, the tunnel barrier can be very thin and the coupling consequently much higher. Here, I report conductance peaks as large as 1500% of background, with values of 300% being typical. Although the nature of the local moment remains in question, I find that the conductance contribution due to the Kondo scattering can actually be reversibly switched on and off by mechanically "squeezing" the junction.

The conventional theoretical interpretation of zero-bias peaks is as follows: Appelbaum [3] treated tunneling in the presence of a local moment in the barrier in the transfer-matrix formalism, introducing an *s-d* exchange interaction term into the Hamiltonian. This interaction is the same as that responsible for the Kondo effect in dilute alloys of transition metals in noble metals. By analogy to Kondo's perturbation treatment of magnetic alloys [5], Appelbaum showed that third-order perturbation theory gave a contribution to the tunnel conductance displaying the behavior seen in metal-insulator-metal junctions, that is, a peak logarithmic in both bias and temperature. Appelbaum's perturbation expansion gives essentially three additive contributions to the junction conductance: $G = G_1 + G_2 + G_3$, where $G_1$ contains all the nonexchange tunneling contributions, including spin-independent scattering by the local moment. In zero applied field and for the case where only a single local moment is present,

$$G_2 = (8\pi^2 e^2/h)\rho_A(E_F)\rho_B(E_F)S(S+1)T_{J_A}^2 \tag{1}$$

and

$$G_3 = 4G_2 J_A \rho_A F(eV) , \tag{2}$$

where $\rho_A(E_F)$ and $\rho_B(E_F)$ are densities of states of conduction electrons in the electrodes $A$ and $B$ at the Fermi energy, $S$ is the spin on the local moment, $J_A$ is the exchange interaction between an electron in electrode $A$ and the local moment, and $T_{J_A}$ is the exchange tunneling interaction in which an electron tunnels from $A$ to $B$ while exchanging spin with the local moment. In Eq. (2)

$$F(eV) \approx \ln[|eV| + nk_B T/E_0] \tag{3}$$

is an approximation to the Kondo integral, where $n \approx 1.4$ and $E_0$ is an energy cutoff that appears as a fitting parameter.

$G_2$ is a second-order perturbation that corresponds to spin-flip tunneling. In zero field it gives a voltage-independent positive contribution to the conductance. $G_3$ appears in third-order perturbation treatment and gives a logarithmic peak in zero field. Immediately, one recognizes that, for perturbation theory to be applicable, $G_3/G_2 \ll 1$. This condition has always been satisfied in previous experiments on conventional tunnel junctions, where one would have needed to reach extremely low temperatures before the divergence of Eq. (3), and thence of Eq. (2), would cause the perturbation expansion to be invalid. Breakdown of the expansion is more than just an annoyance; in the conventional treatment of the Kondo problem it is believed that this situation would be accompanied by the onset of spin correlations. In the present experiment the condition $G_3/G_2 \ll 1$ is *rarely* met. Consequently, the perturbation treatment of Appelbaum should generally *not* be valid. Instead, a nonperturbative approach, such as that of Appelbaum, Phillips, and Tzouras [6] is required. These authors find that, in the strong-coupling case, the conductance peak should remain logarithmic in both temperature and bias, but saturate in the limit of very strong coupling. They point out, however, that their conclusions are tentative, being based on a rather inadequate solution for the Kondo ground state.

The crossed-wire tunnel junction technique has been described elsewhere [7]. It produces junctions in which tunneling occurs between naturally occurring atomic-scale asperities on the wires. These junctions are therefore truly microscopic. Here, I use 99.95%-purity 10-$\mu$m-diam tungsten wires that can be Ohmically heated to a temperature high enough to remove all surface contamination, as indicated by the fact that contacting wires cold weld after cleaning. Wire spacing is controlled by passing a dc current through one of them in a perpendicular magnetic field. The tunneling barrier is formed by condensing a helium film on the wires and bringing them together. Perhaps surprisingly, this helium layer also gives considerable mechanical stability to the junction. In fact, I have discovered that, if helium-coated wires are "slapped" together the deflecting current can be removed, thus allowing arbitrarily large magnetic fields to be applied without inadvertently altering the junction geometry.

The first question to answer is whether these junctions can reproduce the kind of behavior that is seen in conventional junctions in finite magnetic fields. This should occur when the conductance peak is relatively small, in which case the Appelbaum [3] approach should be valid. The following behavior should be observed: A magnetic field should change the $G_2$ contribution from elastic to inelastic, generating a "notch" of width $2g\mu_B H$, where $g$ is the $g$ factor of the local moment, $\mu_B$ the Bohr magneton, and $H$ the applied field. The $G_3$ peak should split into three peaks separated from each other by $g\mu_B H$. In Fig. 1 I show data corresponding to these conditions. The conductance peak develops a dip in a manner very similar to that of peaks observed by Shen and Rowell [8]. These authors analyzed their data by assuming that the $G_3$ peak did not, in fact, split and fitted the $G_2$ term with a rounded V-shaped notch. However, later experiments by other workers using higher fields and lower temperatures [9,10] seemed to require split $G_3$ peaks in addition to the $G_2$ dip. The data of Fig. 1 do not appear to indicate a splitting of the $G_3$ peak, in agreement with the observations of Shen and Rowell [8]. Because the data of Fig. 1 are smeared considerably ($\approx 1$ mV) at the measurement temperature of 3.5 K, no attempt will be made here to extract a value for the $g$ factor.

For the remainder of this paper, after subtracting the typical shallow parabolic conductance background, I use the peak height in zero magnetic field at a temperature of 1.5 K as the $G_3$ characterizing a junction. I take the reduction in peak height in a field of 5.0 T at 1.5 K as the measure of $G_2$. Owing to temperature smearing, the reduction associated with $G_2$ is not fully developed, so the procedure is not entirely accurate, but it enables one to make comparisons between peaks obtained under the same conditions of magnetic field and temperature. The quantity to be used in such comparisons is "$G_3/G_2$" where the quotes emphasize that approximation is involved.

I now consider the new regime where conductance peaks much larger than those of Fig. 1 are observed. One such peak is plotted in Fig. 2. The inset shows that the peak is hardly affected by a field of 5.0 T, which by the above procedure allows one to estimate "$G_3/G_2$" $\approx 43$. Although this ratio has limited quantitative significance, it clearly indicates that this junction is likely to be in the strong-coupling regime. To be more concrete, the Kondo temperature $T_K$ has been defined in connection with the dilute magnetic alloys (see, for instance, Ref. [11]) as the characteristic temperature below which the system is

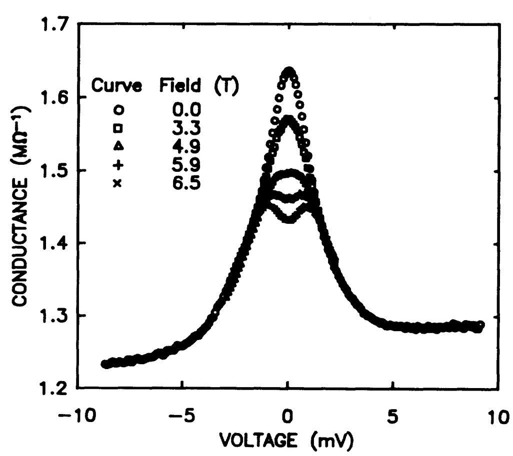


FIG. 1. Dependence of the small-bias conductance on magnetic field for a junction with "$G_3/G_2$" $\approx 0.3$. Measurement temperature is 3.5 K.

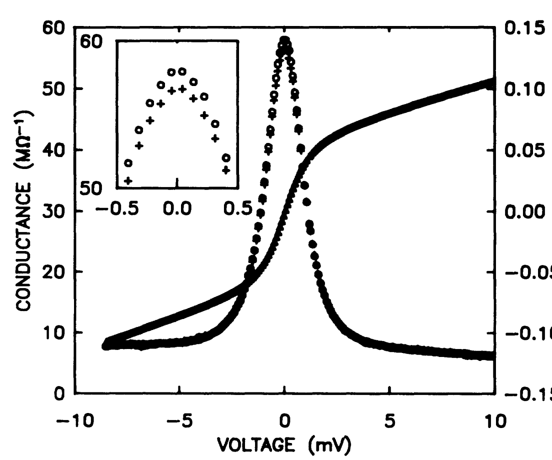


FIG. 2. $I$-$V$ curve and conductance feature for a junction with "$G_3/G_2$" $\approx 43$ in zero field (open circles and triangles) and conductance in 5.0-T magnetic field (crosses). Inset: Conductance data at peak. Measurement temperature is 1.5 K.

strongly coupled. It is given by the well-known expression

$$T_K \approx T_M e^{-1/J\rho_A}, \tag{4}$$

where $T_M$ characterizes the magnetic interaction. $J\rho_A$ can be estimated from Eqs. (2) and (3). The cutoff energy $E_0$ in Eq. (3) has been found to be about 10 meV by earlier workers [8,10,12,13] from the dependence of peak height on the logarithm of bias voltage. The present data give $E_0 \approx 5$ meV. Using this value and $T=1.5$ K in Eq. (3) implies that $J\rho_A \approx 3.6$"$G_3/G_2$". Thus, if "$G_3/G_2$" $>1$, $T_K \gtrsim 0.8 T_M$. Now, consider the data of Fig. 3. If $T_M > 16$ K, as seems likely, then $T_K$ is higher than all the temperatures plotted in Fig. 3. The data in this figure correspond to the strongly coupled regime and do, in fact, fall on a universal straight line. As suggested by Ref. [6], the logarithmic temperature dependence of Eq. (3) clearly remains valid.

Having established that strong exchange scattering is present in the tungsten junctions, is there more that we can say concerning the local moments themselves? Consider the $I$-$V$ curves in Fig. 4(a) in which the conductance is switching between two states. One of the states displays the strong nonlinearity caused by exchange scattering and the other does not. By adjusting the deflecting force between the wires, and thereby the stress in the junction, a condition is found in which a small increase or decrease puts the junction reversibly into the other state. When precisely adjusted, the junction spontaneously and randomly switches between the two states. Because the background conduction differs only somewhat between the two $I$-$V$ curves, despite an exponential dependence of the conductance on barrier width, it would seem unlikely that the conduction path is switching between two physically distinct regions. Therefore, let us consider the proposition that, in a single microscopic junction, a single local moment is responsible for the scattering and this local moment either appears and disappears or greatly changes its coupling to the electrodes. Figure 4(a) was obtained by applying a very small periodic modulation to the deflecting force, illustrating that this type of junction acts as a tunable magnetic scatterer.

In Fig. 4(a) it can be seen that the background linear junction conductance is higher in the state with stronger exchange scattering. This is to be expected because the $G_2$ contribution, which is linear, is also switching. I now consider possible explanations of the observed switching. Planar metal/oxide/metal tunnel junctions commonly display switching between linear conductance states, although it is usually a very small effect. It is also seen in crossed-wire junctions with various wire materials [e.g., Fig. 4(b)] and $\Delta G/G$ can range up to unity. This purely linear conductance switching can also be tuned. The effect is believed due to the presence of electron traps in the tunnel barrier [14]. In Ref. [14] it is suggested that

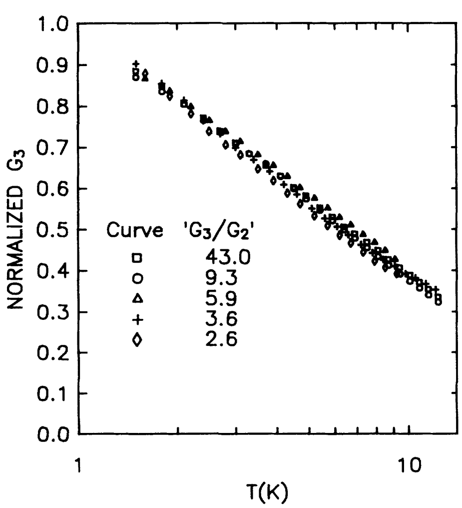


FIG. 3. Temperature dependence of the zero-field value of $G_3$ for junctions with "$G_3/G_2$" ratios as indicated. The conductance is normalized to unity at a temperature of 1.0 K.

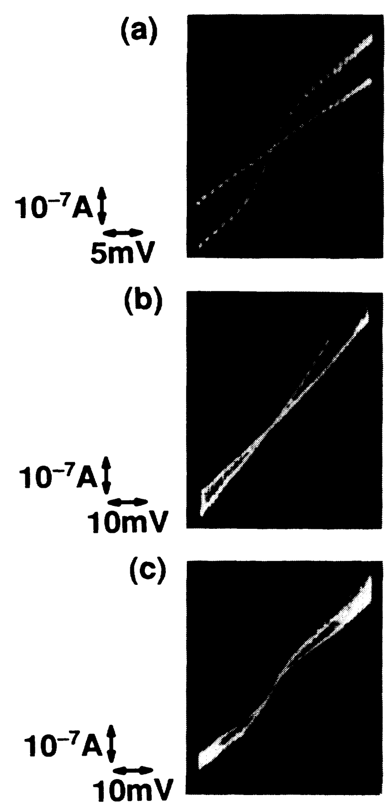


FIG. 4. (a) Switching behavior of the $I$-$V$ curve for a tungsten junction at 1.5 K. A magnetic field of 3.3 T and a combined dc and ac deflecting current is applied to put the junction into a driven switching state. (b) Spontaneous switching of purely linear conductance in a tungsten junction at 1.5 K. (Absence of any evidence of magnetic scattering in a tungsten junction is quite a rarity.) (c) Spontaneous switching between junction states with different but finite magnetic scattering.

the traps are, in fact, microscopic systems in which atomic rearrangement causes trapping and ejection of electrons. These systems most commonly have two available configurations, associated with which are two coupled potential wells. Transitions between the configurations will be thermally activated at high temperatures and at low temperatures involve atomic tunneling. This is a random process and the resulting charge fluctuation modulates the tunnel barrier. Hence the conductance displays "telegraph" or "switching" noise.

It might seem straightforward at this point to make the association between electron traps and the local moments giving rise to exchange scattering. That is, a trap either possesses a magnetic moment when filled and not when empty, or vice versa. However, while an electron trap should certainly have a magnetic moment, it is an experimental fact that junctions formed between gold or platinum wires display switching between linear conductance states but no evidence of a zero-bias Kondo scattering feature. Therefore, it seems that either a different kind of local moment is also present in the tungsten junctions or that the coupling of the trap moments to the electrons in the electrodes is much stronger for tungsten.

Consider the first possibility: At this point only three wire materials have been studied, so it is likely that other materials will display the behavior reported here for tungsten. Perhaps the crucial difference between tungsten and gold or platinum is that the surface of the tungsten wires may have quasi-isolated atoms, either because of local geometry or because some small amount of tungsten oxide remains after the cleaning steps. With its $5d^4$ electron configuration, the tungsten atom has excited states with total angular momentum 1 or 2 when located in a crystal field strong enough to quench its orbital angular momentum. I speculate that the extremely large current density in the junction might pump the excited states. From Eqs. (1) and (2) it can be seen that, if the exchange couplings remain unchanged, then for $S=0$, 1, and 2, $G_3 \propto 0$, 1, and 3. In fact it is observed that switching Kondo peaks do frequently change height by a factor of about 3. This is the case for the $I$-$V$ curves of Fig. 4(c).

The second possibility is that the local moment is indeed associated with a configurational trap; but now suppose that the electron is not ejected, but simply moves between the potential wells. Whereas switching between linear conductances is largest when a trap is at the center of the junction, Kondo scattering is largest when the local moment is near an electrode and can couple strongly to the conduction electrons. Thus, a trap right at the metal surface would be a strong Kondo scatterer and the scattering strength would switch as the moment switched position from one side of the trap to the other—hence the adjustability under junction stress.

To summarize, while the explanations of the switchable moments are hypothetical, these moments certainly give rise to conductance enhancements 2 orders of magnitude larger than previously observed. This indicates that the junctions are in the strong-coupling regime, which has not been attained previously but which can be studied in a controllable way by the present technique.

I would like to thank C. T. Rogers for helpful discussions.

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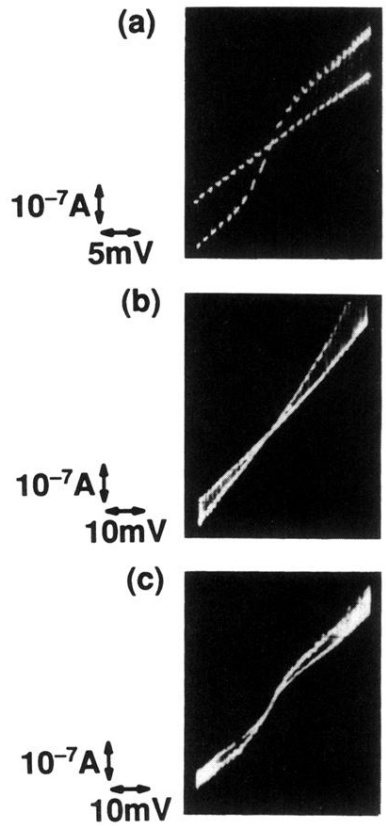


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