

## Observation of Coulomb Correlations of Resonant Tunneling and Inelastic Hopping

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We have observed Coulomb correlations between the two resonant tunneling conduction channels, one per spin orientation, associated with each localized site in the barrier of a metal-insulator-metal tunnel junction. Our data support the recent theory of Glazman and Matveev pertaining to correlated resonant tunneling. We also infer correlated inelastic hopping along chains consisting of two localized sites.

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The process of resonant tunneling via localized states or laterally confined structures is of central importance in many physical systems attracting current attention, including double barrier resonant heterostructure devices [1,2], mesoscopic structures incorporating a two-dimensional electron gas [3,4], amorphous silicon tunnel junctions [5-7], and novel Josephson junction systems [7,8]. In the single electron picture, the total resonant current is simply the sum of the currents passing through each individual resonant channel. In principle, two electrons with opposite spins can occupy each resonant site, and thus each localized site gives rise to two resonant channels. The single electron picture is not a good approximation, however, when there is a large on-site Coulomb interaction,  $U$ , and the tunneling becomes correlated. Such correlated tunneling has recently generated much theoretical [9] and experimental [2,4] interest. In this Letter, we report the observation of correlated tunneling in a Mo/*a*-Si/Mo tunnel junction. Our results are consistent with the recent predictions of Glazman and Matveev [10], and further demonstrate the presence of correlations in inelastic hopping transport channels involving multiple localized sites. The latter effect may have a wider application to the role of Coulomb effects in variable range hopping, which recently has acquired renewed interest [11].

As we have shown previously, amorphous silicon tunnel barriers constitute an effective model system for studying tunneling via localized states [5,6]. Three features of this system greatly simplify the calculation of the correlation effects, making *a*-Si uniquely suitable for an unambiguous test of the theory. First, each localized site (arising in our case from an uncoordinated Si-Si bond) is essentially a delta-function potential well within the barrier and has only *one* (discrete) bound energy level. In a sense, each site can be thought of as a quantum dot in the limit of extreme confinement, giving rise to just two localized states, one per spin orientation. Second, these states are distributed evenly and densely throughout the barrier both in space and in energy sufficiently close to the Fermi level. Third, the localization of the wave functions of tunneling electrons within  $\alpha^{-1}$  ( $\approx 7$  Å) of a resonant center leads to a Coulomb energy for the simultaneous resonant tunneling of two electrons via a single site,  $U$ , on

the order of 100 meV. Therefore, the two resonant channels through a single site are in general highly correlated; while an electron is tunneling through a particular localized state, it blocks resonant transport through the other state associated with the same site. The degree of correlation for each site depends upon its average occupation, which in turn depends upon its position in the barrier.

Also, singly occupied sites that have an energy close to  $\epsilon_F - U$  participate in resonant tunneling due to the on-site Coulomb interaction, since the addition of a second electron to these sites from the Fermi surface of the electrodes is elastic. These low-lying sites manifest the same correlations as the sites near the Fermi level, as is easily seen by considering holes tunneling through them in the reverse direction: A hole tunneling via a low-lying localized state blocks the other channel associated with the same site, since two holes cannot occupy one of these low-lying sites simultaneously. A low-lying site yields the same contribution to the resonant conductance as a site near the Fermi level at the same location.

Glazman and Matveev [10] have calculated the resonant tunneling conductance under the conditions  $U \gg kT \gg eV$  using kinetic equations, which are valid in the limit  $T \gg T_K$ . (The resonant tunneling Kondo temperature  $T_K$  is less than 1 mK for our system [12].) Since the density of localized sites is large and their configuration is fixed in our junctions, the most accessible test for the presence of Coulomb correlations is the consequent characteristic dependence of the zero bias conductance upon an applied magnetic field. Glazman and Matveev show that when a strong magnetic field ( $B \gg kT/\mu_B$ ) lifts the degeneracy of the two spin states associated with each localized site, the result is that all of the resonant tunneling occurs through uncorrelated channels. The correlations vanish in the presence of a strong field because at most only one spin state associated with each site can be close enough to the Fermi level in the electrodes to contribute to the zero bias conductance.

Thus, for a uniform distribution of localized sites, the ratio of the resonant conductance in a strong field to that in zero field directly reflects the degree of the correlation of the two channels linked to every site, and is a universal number. For intermediate fields, Glazman and Matveev predict that the zero bias resonant tunneling conductance

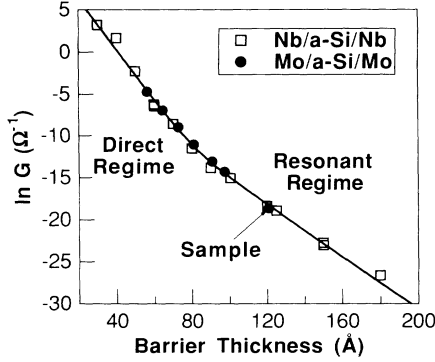


FIG. 1. The logarithm of the zero bias conductance (normalized to a junction area of  $8 \times 8 \mu\text{m}^2$ ) plotted against barrier thickness for a series of junctions, showing the crossover from predominantly direct to predominantly resonant tunneling. The solid line is a fit by the form  $\ln G = \ln(G_{\text{dir}} e^{-2ad} + G_{\text{res}} e^{-ad}/d)$  with  $G_{\text{dir}}$ ,  $G_{\text{res}}$ , and  $a$  as free parameters, yielding  $a = 6.8 \text{ \AA}$ .

$G_R$  obeys a universal dependence on the ratio of the Zeeman energy of the electrons to the thermal broadening of the Fermi surfaces in the electrodes,  $x = \mu_B B/kT$ . After averaging over a uniform distribution of localized states in the barrier, they obtain

$$\frac{G_R(B)}{G_R(0)} = F_R(\mu_B B/kT) = \frac{e^{2x} \ln(1 + e^{-2x}) + e^{-2x} \ln(1 + e^{2x})}{2 \ln 2}. \quad (1)$$

Note that  $G_R(\infty)/G_R(0) = (2 \ln 2)^{-1} \approx 0.721$ .

We have fabricated and measured over 100 Mo/a-Si/Mo and Nb/a-Si/Nb tunnel junctions in the course of our investigation of the role of localized states and barrier interactions in tunneling. Depending upon the thickness of the barrier, the temperature, and the bias voltage, we have observed direct and resonant tunneling, inelastic hopping along chains of localized states, variable range hopping, and Kondo-like zero bias anomalies in the direct and perhaps the resonant tunneling [5,6]. Below we present zero bias conductance data, taken using a lock-in technique with an excitation bias of less than  $20 \mu\text{V}$ , from a single Mo/a-Si/Mo junction with a barrier thickness of  $120 \text{ \AA}$  and an area of  $90 \times 90 \mu\text{m}^2$ . Junctions with thicker barriers have too high a resistance to measure with the necessary accuracy and are dominated by inelastic hopping, while direct tunneling and Kondo-like zero bias anomalies interfere in junctions with thinner barriers. Figure 1 shows the crossover as a function of thickness from direct to resonant tunneling and indicates the sample under discussion; from this graph we infer that direct tunneling comprises less than  $\frac{1}{2}\%$  of the conductance of our sample, so we can safely neglect it.

In Fig. 2 we plot the zero bias conductance of our sample for four different temperatures as a function of the applied magnetic field up to 10 T. The measurement is

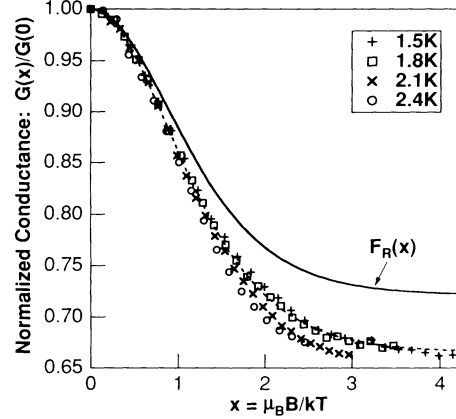


FIG. 2. The normalized magnetic field dependence of the zero bias conductance of the sample, plotted as a function of the parameter  $x$ , for four temperatures, demonstrating universal behavior. The solid line shows the theoretical prediction, while the dashed line shows the theory scaled to give 20% more suppression than predicted.

insensitive to the orientation of the field, confirming that its effect upon the orbital motion of the tunneling electrons is negligible. The ordinate of each curve is normalized to the value of the conductance at that temperature in zero field and the abscissa is scaled to  $x = \mu_B B/kT$  for each curve. (The actual resistance is about  $1 \text{ M}\Omega$  at 4.2 K. We fabricated and measured a second junction nominally identical to the first to confirm these results and found the same behavior.) Independent of any theoretical prediction, the universal behavior of  $G$  as a function of  $x$  demonstrates directly that we are seeing a pure electron spin effect. Note that the deviations at large  $x$  are systematic and increase with increasing temperature. The solid line shows the prediction of Glazman and Matveev for the case of resonant tunneling, in which there are no adjustable parameters. The suppression of the conductance appears to be 20% greater than predicted, as the dashed line indicates.

That the theory slightly underestimates the magnitude of the observed suppression is not unexpected, since a non-negligible component of the conductance is due to inelastic hopping rather than resonant tunneling, even at 1.5 K. Figure 3 shows the temperature dependence of the zero bias conductance in zero field for the same junction shown in Fig. 2. Below 7 K, the sole appreciable inelastic contribution is from processes of the type depicted in the inset of Fig. 3 [6]. An electron tunnels from the left electrode to the first site, hops inelastically to the second site accompanied by the absorption or emission of a phonon, and finally tunnels to the right electrode. Glazman and Matveev [13] have also calculated the zero bias conductance due to inelastic channels of this type,  $G_2$ , and they found that  $G_2(T) = \sigma_2 T^{4/3}$  for  $eV \ll kT$ . We have verified this simple power-law dependence for a wide variety of sample thicknesses and have also found the

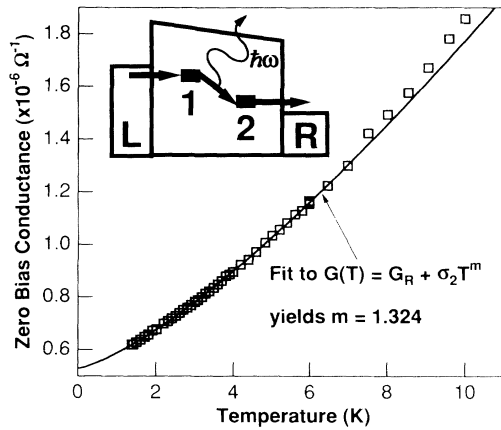


FIG. 3. Temperature dependence of the zero bias conductance, in excellent agreement with the theory of two-site inelastic hopping, which predicts  $m = \frac{4}{3}$  (Ref. [13]). Inset: Schematic representation of two-site inelastic hopping conduction (see text).

complicated prefactor to be in excellent agreement with the theory [6].

The solid line in Fig. 3 is a fit to the form  $G(T) = G_R + \sigma_2 T^m$ , with  $G_R$ ,  $\sigma_2$ , and  $m$  as free parameters. The best fit from 1.4 to 7 K yields  $m = 1.324$  (theory predicts  $\frac{4}{3}$ ); the onset of hopping channels consisting of three localized states causes the deviation above 7 K. Since the temperature dependence of the resonant tunneling [14] is completely negligible here, we can use the fit to calculate the fraction of the conductance at each temperature due to resonant tunneling. The values range from 84% at 1.5 K down to 74% at 2.4 K and are shown in the inset of Fig. 4 [15].

In order to proceed, we now assume that Eq. (1) correctly predicts the correlated tunneling dependence of the resonant component of the conductance. We can then empirically determine the magnetic field dependence of the two-site inelastic hopping channel,  $G_2(B, T)$ , by subtracting the assumed field dependence of the resonant component of the conductance, weighted by the fraction of the conductance due to resonant tunneling, from each of the curves in Fig. 2. That is, equivalently,

$$G_2(B, T) \equiv G(B, T) - G(0, 0) F_R(\mu_B B / kT).$$

Here  $G(B, T)$  is the measured zero bias conductance, which includes both resonant and inelastic processes, and  $G(0, 0)$  is the same as  $G_R$  found in Fig. 3. Figure 4 shows the resulting  $G_2(B, T)$  curves, plotted against  $x = \mu_B B / kT$ , after the renormalization of each curve to its value at  $x = 0$ .

Note that the inelastic channel also exhibits a universal dependence on the parameter  $x$ , which we denote  $F_2(x)$ . Moreover, the deviations at large  $x$  are not systematic in the temperature here, as they are in Fig. 2; we believe that they represent the experimental limitations of our

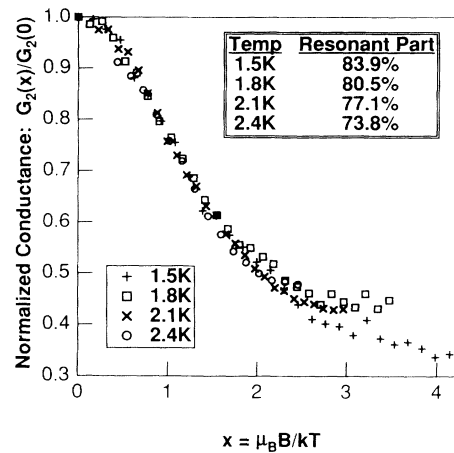


FIG. 4. The magnetic field dependence of the inelastic hopping conduction, extracted from the data as described in the text, plotted as in Fig. 2. Inset: The fraction of the total conductance at each temperature due to resonant tunneling.

measurements. Note also that the inferred suppression is roughly twice as strong for the  $G_2$  channel as for the resonant channel. Such behavior is reasonable since kinetic equations also govern the  $G_2$  channel, but the Coulomb effects apply on the two sites that comprise each inelastic channel and possibly also between the two sites, so the overall correlation is stronger. Unfortunately, there is so far no quantitative prediction of the functional form  $F_2(x) = G_2(B, T) / G_2(0, T)$  with which to compare our results. Nevertheless, the empirical extraction of universal behavior along with the reasonable value  $G_2(\infty) / G_2(0) \approx 0.4$  gives us confidence that Glazman and Matveev's prediction for the resonant case [10] is in fact valid.

One reason that  $F_2(x)$  is worth understanding theoretically and experimentally is that, in contrast to  $F_R(x)$ , it may depend upon whether the localized states that play a role in conduction are actually at the Fermi level, singly occupied and at an energy  $U$  below the Fermi level, or a combination of these two possibilities in some definite proportion. This distinction may, in turn, have important consequences not only for inelastic hopping across thin barriers, but also for the unresolved issue of the effect of Coulomb interactions on variable range hopping.

In summary, we have demonstrated Coulomb correlations between the spin-down and spin-up resonant tunneling channels that share the same localized site and we infer good agreement with the universal dependence predicted by Glazman and Matveev [10]. We have also observed these correlations in more complicated conduction processes involving electron-phonon interactions and found that they too obey a universal dependence.

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