

### BRIEF REPORTS

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#### Low-energy restoration of Fermi-liquid behavior for two-channel Kondo scattering

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The origin of zero-bias anomalies (ZBA's) in pure metal nanoconstrictions is investigated. It is shown that the ZBA's in titanium nanoconstrictions can be completely understood in terms of electron-assisted tunneling systems. The behavior of such a system is expected to depend on two distinct energy scales, a Kondo energy below which the electrons behave in a non-Fermi-liquid manner, and a splitting energy below which the Fermi-liquid behavior is restored. Titanium nanoconstrictions have been found to exhibit zero-bias anomalies that show both the non-Fermi-liquid behavior and the low-energy restoration of the Fermi liquid. [S0163-1829(97)02343-6]

Transport experiments on nanometer-size metallic structures have revealed new physics not readily resolvable in the macroscopic limit. One of the more intriguing features of such nanoconstrictions and point contacts is that they sometimes show zero-bias anomalies<sup>1-3</sup> (ZBA's) in the voltage-dependent conductance  $G(V)$  which cannot be satisfactorily explained by the standard picture of Kondo scattering by magnetic impurities.<sup>4</sup> There are three competing theories which attempt to explain such ZBA's. In the theory proposed by Wingreen, Altshuler, and Meir (WAM),<sup>5</sup> the anomalies are caused by an electron-electron interaction induced reduction in the single-electron density of states in the disordered regions of the nanoconstriction. In the Kozub-Kulik (KK) theory,<sup>6</sup> the nonequilibrium electrons change the occupation levels of slow two-level tunneling systems (TLS) giving rise to a voltage dependent conductance. In the Vladar and Zawadowski (VZ) theory,<sup>7</sup> electron-assisted tunneling between the two positions of a TLS causes non-Fermi-liquid behavior of electrons in the vicinity of the TLS, with the ZBA behavior determined by the two-channel Kondo (2CK) fixed point.<sup>8</sup> The predictions of the three theories differ in important ways: KK predict  $G(V) \approx V^{2/3}$  for high energies (bias voltages) and a saturation in  $G(V)$  as  $V \rightarrow 0$ ; the WAM theory predicts  $G(V) \approx V^{1/2}$  but no saturation at low energies beyond thermal smearing; the VZ/2CK theory predicts that between a Kondo energy and a splitting energy,  $G(V) \approx V^{1/2}$ ,

but that below a crossover energy  $V_x$  the conductance should saturate due to a nonzero splitting between the two lowest-energy levels of the TLS.

Previous experiments of Ralph *et al.* with copper nanoconstrictions<sup>1</sup> observed ZBA's which scaled in a manner consistent with an exponent of  $0.50 \pm 0.05$  but there are aspects to those measurements that cannot be explained by the VZ/2CK model. First, there was no indication of a breakdown of the two-channel Kondo scaling due to the expected splitting of the TLS, down to the lowest temperatures examined,  $T \geq 125$  mK. In addition, strong, "critical bias" conductance features were observed in the Cu nanoconstrictions at high biases, and the ZBA's had a very strong magnetic-field ( $H$ ) dependence. In the experiments of Akimenko and Gudimenko<sup>3</sup> with Cu mechanical point contacts, and of Keijsers, Shklyarevskii, and Kempen<sup>2</sup> with mechanically controlled break junctions made from Cu, Au, Ag, and Pt, ZBA's of both signs were seen, consistent with the KK theory. More recent experiments of Keijsers, Shklyarevskii, and van Kempen<sup>2</sup> on metallic glasses show very clearly that the anomalies arise from two-level systems but they cannot determine whether or not *electron-assisted* tunneling is important.

In this paper we present results of an experiment designed to establish the origin of the zero-bias anomalies in pure metal point contacts. We show that in titanium nanoconstrictions, the zero-bias anomalies are completely consistent with

the existence of electron-assisted tunneling in two-level systems. In particular, these ZBA's exhibit precise scaling behavior with an exponent of  $0.52 \pm 0.05$  that is consistent with the WAM and VZ/2CK theories but not with KK, and a saturation behavior that is consistent with KK and VZ/2CK but not with WAM. We also find that it is possible to change the low-energy behavior of  $G(V)$ , via short-range electromigration, without changing the high-energy behavior. This result is readily explainable in the context of VZ/2CK theory, but not with electron-electron-scattering effects. Finally, we find that the magnetic-field ( $H$ ) dependence of these zero-bias anomalies is very weak and limited to the low-energy part of the ZBA's, which can be interpreted as due to a change in the splitting energy by the rearrangement of phases along the Feynman paths,<sup>9</sup> but which is inconsistent with the WAM and KK theories. Thus the ZBA behavior of these titanium nanoconstrictions is fully consistent only with the predictions of the VZ/2CK theory. Since they do not exhibit the additional features observed in the Cu nanoconstrictions, these nanoconstrictions open the possibility of studying the physics of the "clean" 2CK model in detail, as well as allowing the examination of a system that shows a crossover from, two-channel Kondo to a Fermi-liquid behavior.

Point-contact spectroscopy has been used for some time to study energy-dependent scattering<sup>10,11</sup> of electrons. For a contact diameter between two electrodes that is shorter than the low- $T$  mean inelastic-scattering length of the material, it is possible to drive the Fermi sea near the contact out of equilibrium by an amount  $\sim eV$ , where  $V$  is the bias applied across the device. The change in conductance  $G$  as a function of  $V$  is then a measure of the energy- (or equivalently, temperature-) dependent scattering rate. This effectively allows the determination of the  $T$  dependence<sup>10</sup> of the zero-bias scattering rate by a measurement of  $G(V)$  at a very low, fixed  $T$ .

For this study we fabricated Ti nanoconstrictions using nanolithographic techniques discussed elsewhere.<sup>12</sup> In brief, the samples are prepared by electron-beam evaporation in a vacuum of mid  $10^{-8}$  Torr onto both sides of a silicon-nitride membrane containing a nanohole (3–10 nm diameter). We find that these Ti nanoconstrictions are considerably more stressed than similarly prepared Cu devices. Indeed, the tensile stress is sufficient for some devices to pull apart soon after fabrication, or for the device resistance to change spontaneously over several hours, usually in an upward direction, even at very low  $T$ . This high level of tensile stress in these nanoconstrictions makes them likely candidates to have dynamical defects. Moreover, due to the higher melting point of Ti, structural defects in these nanoconstrictions anneal away very slowly during room-temperature storage, as compared to Cu nanoconstrictions.

After preparation, the samples were transferred to a He dilution refrigerator and cooled to 75 mK. The samples were biased with a slowly varying current and a small ac wiggle was used to measure the differential resistance using lock-in detection. Even at the lowest temperatures our excitation voltage (1  $\mu$ V) was less than  $k_B T/e$  (6  $\mu$ V for 70 mK).

More than 90% of the Ti samples that we have studied had a ZBA when they were cooled to low  $T$ . The Fig. 1 inset shows the conductance  $G(V)$  vs  $V$  for a Ti nanoconstriction (sample No. 1) at different temperatures. In Fig. 1 we plot

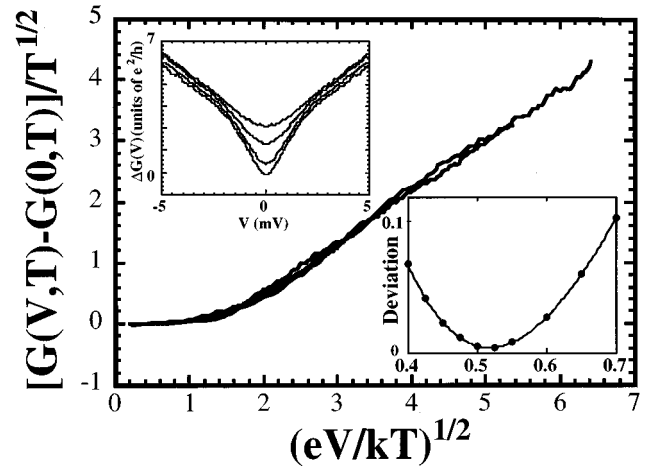


FIG. 1. Bias-dependent conductance in the scaling form  $[G(V,T) - G(0,T)]/T^{1/2}$  vs  $(eV/k_B T)^{1/2}$  for a  $20\Omega$  (sample No. 1) titanium nanobridge at 6.0, 4.0, 2.0 and 1.4 K in zero magnetic field. Top inset:  $\Delta G(V) = G(V,T) - G(0,1.4\text{ K})$  for the same sample at 6.0, 4.0, 2.0, and 1.4 K (top to bottom). Bottom inset: deviation (arb. units) from scaling as a function of the exponent  $\alpha$ .

the same data in the scaling form  $[G(V,T) - G(0,T)]/T^\alpha$  vs  $(eV/k_B T)^\alpha$  with  $\alpha = 0.5$  predicted by conformal field theory (CFT) calculations.<sup>8</sup> The top inset shows the raw data from which the scaling curves were determined, and the bottom inset shows the deviation from scaling for different values of  $\alpha$ . From this curve we determine that  $\alpha = 0.52 \pm 0.05$ . We take this to be strong evidence in favor of a 2CK interpretation over the KK picture where the predicted exponent<sup>6</sup> is  $\frac{2}{3}$ . The 2CK scaling function is characterized by two universal constants,<sup>1,8</sup>  $\Gamma_1$  and  $\Gamma''(0)$ . We have measured these numbers using the method outlined in Ref. 1 and find  $\Gamma_1 = -0.81 \pm 0.10$  and  $\Gamma''(0) \leq 0.1$  for the sample of Fig. 1. The deviation from the CFT prediction ( $\Gamma_1 = -1.14 \pm 0.1$ ) can be explained if corrections of the order  $T/T_K$  are important. Using the NCA (noncrossing approximation) calculations of Hettler, Kroha, and Hershfield,<sup>13</sup> we estimate that  $T/T_K \approx 0.1$ .

Figure 2 shows results from a sample (No. 2) where the scaling breaks down at low temperatures. This is in sharp contrast to the previous Cu nanoconstriction data. The inset shows the  $G(V)$  vs  $V^{1/2}$  for the sample at 1.41 K and 76 mK. For the sample in Fig. 1, the  $V^{1/2}$  behavior sets in at  $eV \approx 2k_B T$ ; this low-voltage saturation is caused by thermal rounding. For the sample in Fig. 2, the low-voltage saturation extends out to  $eV \approx 31k_B T$ . Since this saturation is more than ten times that expected from thermal smearing of the Fermi surface, we infer that this saturation is a true measure of the energy-dependent scattering rate of the TLS. We are certain that the electron temperature is the same as the measured temperature because the same experimental configuration and equipment was used to previously cool Cu nanoconstrictions which exhibit only thermal saturation down to 50 mK, and because here the saturation of the ZBA's can be changed by electromigration.

The observed scaling and its subsequent breakdown finds a natural explanation in the electron-assisted tunneling system picture. When the bias energies of the incident electrons are lower than the splitting between the two lowest energies of the two-level system, the Kondo scattering is cutoff, and

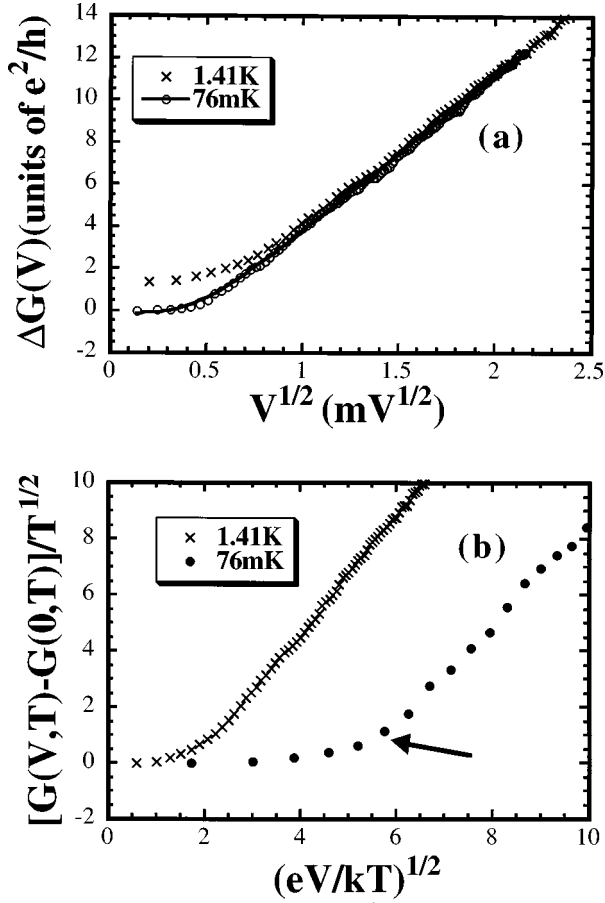


FIG. 2. (a).  $\Delta G(V) = G(V, T) - G(0, 76 \text{ mK})$  vs  $V^{1/2}$  for a  $19\Omega$  titanium nanoconstriction (sample No. 2) at 1.41 K and 76 mK. (b). The same data plotted as  $[G(V, T) - G(0, T)]/T^{1/2}$  vs  $(eV/k_B T)^{1/2}$  at 1.41 K and 76 mK.

the system is restored to a Fermi liquid. From the size of the anomaly  $\approx 10e^2/h$  we can estimate that the scattering is due to about 5 tunneling centers, if we make the standard hypothesis that each tunneling system contributes  $2e^2/h$  to the scattering cross section. Generally, the splittings of the various TLS will be different, but the data suggests that the smallest splitting for the tunneling systems is distinctly nonzero. In order to extract a measure of the mean splitting of the Kondo scatterers for sample No. 2, we fit the 76-mK data in Fig. 2(a) to an empirical form,

$$G(V) = G(0) + a\{(eV)^2 + (2k_B T_x)^2\}^{1/4}, \quad (1)$$

that has the desirable characteristic of a  $\sqrt{V}$  dependence at high bias and a  $V^2$  dependence at low bias. This allows us to estimate the mean crossover temperature  $T_x = 1.43 \pm 0.03 \text{ K}$ . Note, however, that the crossover is rounded, indicative of some spread in the distribution of splitting energies for the small number of TLS in this sample.<sup>7</sup>

The parameters of the tunneling systems should be sensitive to their local environment,<sup>9,14</sup> i.e., to the distribution of other elastic scatterers in their vicinity and to the local atomic strain field. To test the assumption that a level splitting causes the saturation, we examined the effect of rearrangement of the atomic defects. Previous nanoconstriction

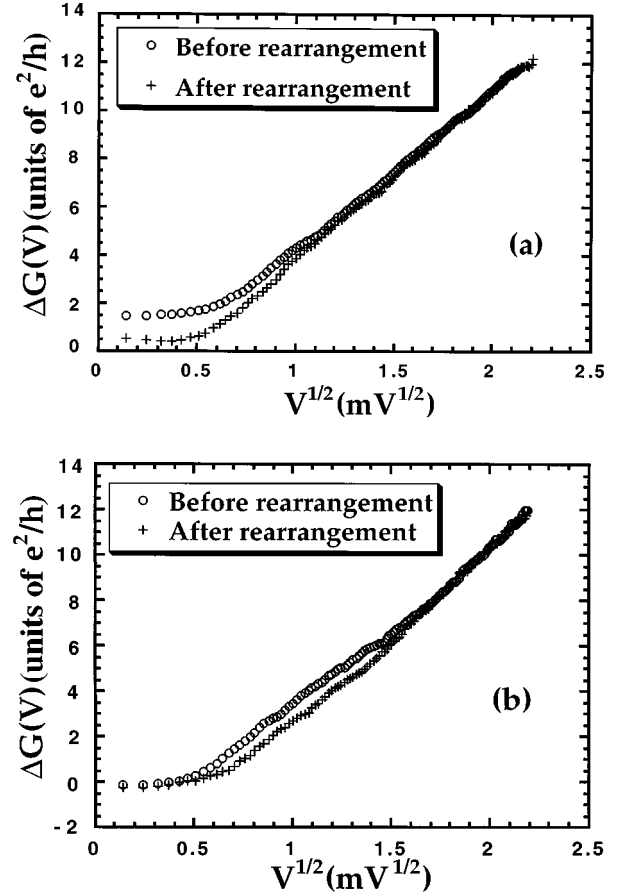


FIG. 3. (a). The low-bias conductance anomaly of sample No. 2 before and after being subjected to a strong electrical bias ( $J \sim 10^{10} \text{ A/cm}^2$ ) for 10 sec. The level splitting of the tunneling system as indicated by the low- $V$  departure from the  $\sqrt{V}$  behavior has been changed. (b) For a  $22\Omega$  device (sample No. 3) the multiple deviations from  $\sqrt{V}$  behavior indicate the presence of tunneling systems with two different splitting energies, which are changed by different amounts upon defect rearrangement by a strong electrical bias.

work has demonstrated that the high current densities (exceeding  $10^{10} \text{ A/cm}^2$ ) can be used to cause defect rearrangement.<sup>15</sup> Sample No. 2 was subjected to a bias of 200 mV ( $J \sim 10^{10} \text{ A/cm}^2$ ) for 10 s several times. Figure 3(a) shows the different low-bias behaviors before and after an electromigration. This change can be interpreted as due to a change in the splitting energy. Using Eq. (1), we estimate that  $T_x$  decreased from 2.3 to 1.4 K due to this defect rearrangement. The observation that electromigration causes a change in the low-bias behavior while the high-bias behavior remains the same is consistent with the existence of two independent energy scales in the scattering process.

In Fig. 3(b) we show  $G(V)$  for another device (sample No. 3) that was changed by the brief application of a very high bias. The low-bias behavior for this sample is not simple, and cannot be described by a single splitting parameter. Before defect rearrangement, the  $G(V)$  characteristic had saturation behavior setting in at two different voltages. Using the method of [Eq. (1)] with the form

$$G(V) = G(0) + a\{(eV)^2 + (2k_B T_{x_1})^2\}^{1/4} + b\{(eV)^2 + (2k_B T_{x_2})^2\}^{1/4}, \quad (2)$$

we find two distinct crossover temperatures of approximately 0.9 and 9.7 K. After rearrangement, the crossover temperatures changed to  $\approx 1.5$  and 6.8 K, respectively. The observation of multiple splittings in the same sample and their distinct behavior under electromigration lends further support to the  $VZ/2$  CK interpretation.

Over the range of the Ti samples that we have studied, we have generally found conductance anomalies consistent with 2CK scattering from  $\sim 2$  to 10 atomic scale TLS defects, but with the somewhat surprising feature that the splitting energies of the TLS defects in a given sample tend to cluster about 1 or more values, resulting in the conductance anomalies of a sample clearly exhibiting one or more crossover temperatures occurring somewhere in the range of  $< 1$  to  $> 10$  K. From sample to sample, however, there appears to be no preferred crossover temperature. This suggests that the TLS splitting energy distribution, when averaged over a number of defect configurations, is relatively broad and flat. The apparent clustering of the splitting energies of the small number of TLS in a particular nanoconstriction around a few values is intriguing and may help identify the microscopic origin of these defects.

Besides the saturation behavior of the ZBA's, the conductance data from these Ti devices differ from those previously obtained with Cu nanoconstrictions in two important aspects. First, the sharp critical bias transitions that were observed in the conductance of the Cu samples<sup>1</sup> have not been observed in any of the Ti samples studied to date. Second, there is little or no magnetic-field dependence to the ZBA behavior of these devices, as seen in Fig. 4(a), which shows  $G(V)$  for a Ti sample as measured in a zero and in a 5-T field. Figure 4(b) shows a different sample where the change is  $\approx e^2/h$ . In the numerous Ti nanoconstrictions that we have studied, the change in  $G(0)$  as a function of  $H$  is  $\approx 2e^2/h$ ; such changes are restricted to the low- $T$ , low- $V$  regime and can be of either sign. It is well known that the splitting energy of two-level systems is sensitive to the magnetic field through the rearrangement of phases along the Feynman paths.<sup>9</sup> The change in only the low-bias behavior with a magnetic field is also consistent with the saturation being caused by the finite splitting of two-level systems.

A magnetic field is expected to quench 2CK scattering at a rate that is linear<sup>8</sup> in  $H$ , but the energy scale for such quenching has not been established. Estimates for the energy

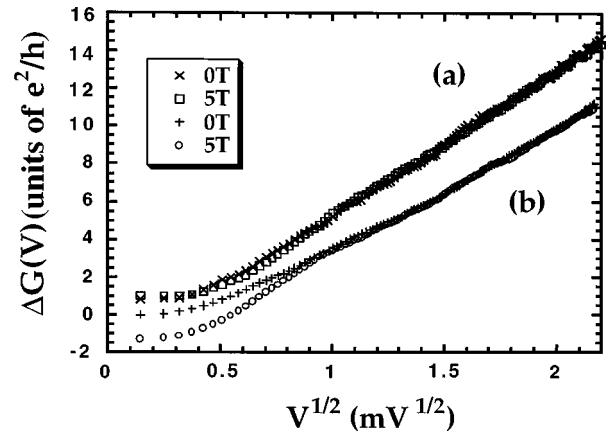


FIG. 4. The magnetic-field dependence (0 and 5 T) at 100 mK for two Ti nanoconstrictions: (a) sample No. 2 (b) sample No. 3. Data for (a) are offset by  $e^2/h$  at zero bias for clarity.

scale required,  $\mu_B H$ , range from  $k_B T_K$  to the Fermi energy  $\epsilon_F$ . The copper nanoconstriction experiments found a strong field effect, with the ZBA shrinking by a factor of two as  $H$  varied from 0 to 2 T. However, we note that in those experiments the field dependence was generally correlated with the reduction to zero  $V$  of the critical bias transition points. The absence from Ti samples of both critical bias transitions and magnetic-field dependence of the ZBA's suggest that both are particular to the Cu samples, perhaps due to the presence of sets of interacting tunneling systems with very low splittings. This then indicates that the intrinsic energy scale for suppression of 2CK scattering from two-level systems is large.

In summary, we have shown data that point unambiguously to the existence of fast electron-assisted tunneling systems. Ti nanoconstrictions show both the scaling behavior consistent with two-channel Kondo physics and the breakdown of scaling consistent with a finite splitting of the TLS's. The magnetic-field behavior is also consistent with the existence of TLS's with a finite splitting. These observations make Ti nanoconstrictions an ideal system for studying the physics of the two-channel Kondo fixed point.

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<sup>1</sup>D. C. Ralph and R. A. Buhrman, Phys. Rev. Lett. **69**, 2118 (1992); D. C. Ralph, A. W. W. Ludwig, Jan von Delft, and R. A. Buhrman, *ibid.* **72**, 1064 (1994); D. C. Ralph and R. A. Buhrman, Phys. Rev. B **51**, 3554 (1995); Jan von Delft *et al.*, Ann. Phys. (to be published).

<sup>2</sup>R. J. P. Keijsers, O. I. Shklyarevskii, and H. van Kempen, Phys. Rev. Lett. **77**, 3411 (1996); Phys. Rev. B **51**, 5628 (1995); Jan von Delft, G. Zarand, and A. Zawadowski, Phys. Rev. Lett. (to be published).

<sup>3</sup>A. I. Akimenko and V. A. Gudimenko, Solid State Commun. **87**, 925 (1993).

<sup>4</sup>M. D. Daybell, in *Magnetism*, edited by G. Rado and H. Suhl (Academic, New York, 1973), Vol. 5.

<sup>5</sup>N. S. Wingreen, B. L. Altshuler, and Y. Meir, Phys. Rev. Lett. **75**, 769 (1995); D. C. Ralph, A. W. W. Ludwig, Jan von Delft, and R. A. Buhrman, *ibid.* **75**, 770 (1995).

<sup>6</sup>V. I. Kozub and I. O. Kulik, Zh. Eksp. Teor. Fiz. **91**, 2243 (1986)

- [Sov. Phys. JETP **64**, 1332 (1986)]; V. I. Kozub, A. M. Rudin, and H. R. Schober, *Solid State Commun.* **95**, 415 (1995).
- <sup>7</sup>A. Zawadowski, *Phys. Rev. Lett.* **45**, 211 (1980); K. Vladar and A. Zawadowski, *Phys. Rev. B* **28**, 1564 (1983); A. Zawadowski and K. Vladar, in *Quantum Tunneling in Condensed Media*, edited by Yu Kagan and A. J. Leggett (Elsevier, New York, 1992), p. 427; A. Muramatsu and F. Guinea, *Phys. Rev. Lett.* **57**, 2337 (1986); P. Nozieres and A. Blandin, *J. Phys. (Paris)* **41**, 193 (1980).
- <sup>8</sup>I. Affleck, *Nucl. Phys. B* **336**, 517 (1990); I. Affleck and A. W. W. Ludwig, *ibid.* **352**, 849 (1991); **360**, 641 (1991); *Phys. Rev. Lett.* **67**, 161 (1991); I. Affleck, A. W. W. Ludwig, H. B. Pang, and D. L. Cox, *Phys. Rev. B* **45**, 7918 (1992).
- <sup>9</sup>B. L. Altshuler and B. Z. Spivak, *Pis'ma Zh. Eksp. Teor. Fiz.* **49**, 671 (1989) [*JETP Lett.* **49**, 772 (1989)]; N. M. Zimmerman, B. Golding, and W. H. Haemmerle, *Phys. Rev. Lett.* **67**, 1332 (1991).
- <sup>10</sup>I. K. Yanson, *Fiz. Nizk. Temp.* **9**, 676 (1983) [*Sov. J. Low Temp. Phys.* **9**, 343 (1983)]; I. K. Yanson, V. V. Fisun, R. Hesper, A. V. Khotkevich, J. M. Krans, J. A. Mydosh, and J. M. Ruitenbeck, *Phys. Rev. Lett.* **74**, 302 (1995).
- <sup>11</sup>A. G. M. Jansen, A. P. van Gelder, and P. Wyder, *J. Phys. C* **13**, 6073 (1980).
- <sup>12</sup>K. S. Ralls, R. A. Buhrman, and R. C. Tiberio, *Appl. Phys. Lett.* **55**, 2459 (1989).
- <sup>13</sup>M. H. Hettler, J. Kroha, and S. Hershfield, *Phys. Rev. Lett.* **73**, 1967 (1994).
- <sup>14</sup>S. N. Coppersmith and B. Golding, *Phys. Rev. B* **47**, 4922 (1993).
- <sup>15</sup>D. C. Ralph, K. S. Ralls, and R. A. Buhrman, *Phys. Rev. Lett.* **70**, 986 (1993).