Temperature dependence of anisotropic magnetoresistance and atomic rearrangements in ferromagnetic metal break junctions

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Recent experiments have found that the anisotropic magnetoresistance (AMR) of nanometer-scale ferromagnetic contacts at low temperature can be larger than that of bulk samples and can exhibit more complicated variations as a function of sample bias and the angle of an applied magnetic field than in the bulk case. Here, we test a proposal that quantum interference of electrons may explain these results, by measuring the temperature dependence of the AMR signals in nanometer-scale contacts made from Permalloy ($Ni_{80}Fe_{20}$), Ni, and Co. We find a strong temperature dependence, in quantitative agreement with expectations for a quantum-interference effect. In the course of making these measurements, we also observed that two-level resistance fluctuations as a function of time, associated with reconfigurations of the atomic structure, are present in all of our samples as the temperature is increased above a few tens of Kelvin, and they can be found in some samples even at 4.2 K. The relative energy of the different atomic configurations can be extremely sensitive to the angle of the sample magnetization, so that the conductance in some samples can be made to change abruptly and reproducibly as the angle of an applied magnetic field is rotated.

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I. INTRODUCTION

Metallic ferromagnets exhibit anisotropic magnetoresistance (AMR)—their resistance changes as a function of the angle θ between the direction of current flow and the sample magnetization. In bulk samples, this is a relatively small effect ($\Delta R/R \sim 1\%$), and in polycrystalline bulk samples, the form of the angular dependence is simply $\Delta R \propto \cos^2\theta$.[1] Recently, several experiments have shown that the AMR can be enhanced in nanometer-scale ferromagnetic contacts at cryogenic temperatures.[2–4] In this case, the resistance variations as a function of the angle of an applied magnetic field can be more complicated than the $\cos^2\theta$ form, and nonmonotonic resistance variations of approximately the same magnitude are also observed as a function of sample bias. A proposed mechanism for the enhanced AMR in nanoscale contacts is that it might be the result of a quantum-interference effect because changing the direction of the sample magnetization may modify the electron orbits traversing the sample due to spin-orbit coupling, thereby reshuffling their quantum interference and changing the resistance.[5,6] Quantum-interference effects are highly sensitive to temperature, so a necessary check on this proposed mechanism is to measure the temperature dependence of the AMR variations. Here, we report that measurements of this temperature dependence are in excellent quantitative agreement with the theoretical expectations.

In the course of exploring the temperature dependent transport properties of the ferromagnetic contacts, we also observed that time-dependent two-level resistance fluctuations (TLFs) become increasingly common in all of our contacts above a few tens of Kelvin and in a few samples are present even at 4.2 K. Similar fluctuations as a function of time have been observed previously in nanometer-scale contacts made from a wide variety of different nonmagnetic metals and have been identified as due to thermally activated reconfigurations of atoms or small groups of atoms within the device.[7–9] What we find in ferromagnetic contacts is that the dynamics of the TLFs can be extremely sensitive to the orientation of the sample magnetization. By changing the angle of an external magnetic field by just a few degrees, it is sometimes possible to force an abrupt, reproducible transition of a TLF between its high and low resistance states. This can produce abrupt steps in sample conductance as a function of field angle that mimic the dependence predicted due to the "ballistic AMR" (or BAMR) effect.[4,10,11] The BAMR theory predicts that changes in the magnetization angle of a nanoscale magnetic contact may produce abrupt changes in conductance due to opening or closing discrete quantum channels in the contact—an intrinsic electronic effect. However, whenever we have observed this sort of abrupt conductance step as a function of field angle, we find that the transitions are associated with structural reconfigurations rather than due to an intrinsic electronic effect alone. Our discussion of these field-angle-induced conductance changes expands upon a previous brief comment,[12] to which there was a response by Sokolov *et al.*[13]

II. TEMPERATURE DEPENDENCE OF ENHANCED ANISOTROPIC MAGNETORESISTANCE

We have used two slightly different sample geometries in our study. In both cases, the ferromagnetic samples are firmly attached to an oxidized silicon substrate in an attempt to minimize potential motion due to magnetostriction and magnetostatic forces, and the devices are defined by electron-beam lithography. One geometry consists initially of a single line of ferromagnetic material (Ni or Co) 200 nm long, 100 nm wide, and 30 nm thick, attached to electrodes that widen gradually [see inset, Fig. 1(a)]. The second sample geometry is the same one used in Ref. 3—initially 30-nm-thick Permalloy ($Py=Ni_{80}Fe_{20}$) in the shape of two elliptical electrodes connected by a 100-nm-wide constriction [see inset, Fig. 1(c)]. Both types of samples are con-

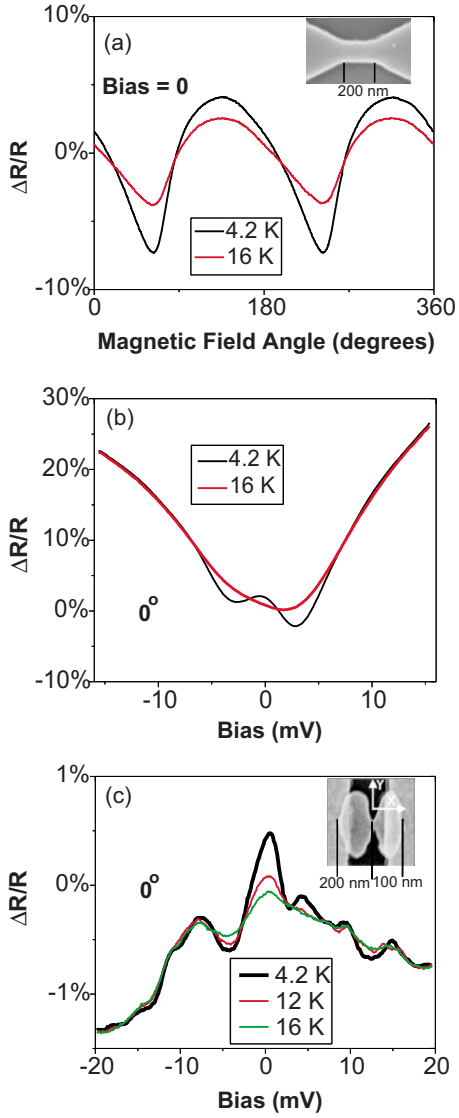

FIG. 1. (Color online) [(a) and (b)] Changes in $R=dV/dI$ for an 8.3 kΩ Ni junction (a) as a function of in-plane magnetic-field angle at zero bias and (b) as a function of bias at a fixed field angle of 0°. (c) Changes in R as a function of bias for a 2.2 kΩ Permalloy junction. The insets show scanning-electron micrographs of the two electrode geometries that we employed. The field magnitude for all traces is 0.8 T, and the traces are normalized by the differential resistance at 4.2 K averaged over a 360° magnetic field sweep. The current direction before electromigration corresponds to a magnetic field angle of approximately 110°.

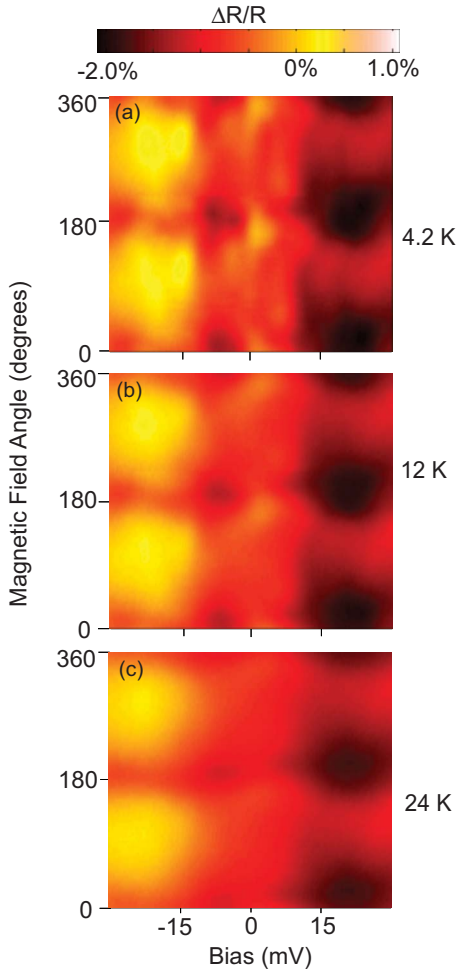

FIG. 2. (Color online) Resistance fluctuations, with magnitude given by the scale at the top, plotted as a function of bias and magnetic-field angle for a 4.05 kΩ Permalloy junction at three different temperatures: (a) 4.2 K, (b) 12 K, and (c) 24 K. The field magnitude is 0.8 T.

nected to larger gold contact pads. To reduce the cross section of the magnetic contacts to a nanometer scale, we mount a sample chip in the vacuum can of a liquid-He "dipstick-style" cryostat and use actively controlled electromigration at a background temperature of $T=4.2$ K.[14] This allows us to narrow each wire gradually and stop the process when the resistance of the sample has grown near to a desired value. We then measure at various temperatures the differential resistance of the sample $R=dV/dI$ as a function of bias voltage and the magnitude and angle of an applied magnetic field. The differential resistance is determined using a lock-in amplifier with an excitation bias small enough not to broaden any features in the data. The temperature is controlled using resistive heating with feedback. We apply the magnetic field with a three-coil vector magnet capable of 0.9 T in any direction. In studies as a function of field angle, we typically apply 0.8 T in the plane of the sample, which is sufficient to fully saturate the sample magnetization.[3]

When any of our devices is narrowed using electromigration to the point that the resistance is greater than about 1 kΩ, we observe enhanced AMR variations at $T=4.2$ K as a function of field angle and as a function of bias, in agreement with the previous study of Py contacts.[3] We find a qualitatively similar behavior in Ni [Figs. 1(a) and 1(b)], Py [Figs. 1(c) and 2], and Co (not shown) samples. When we increase the device temperature from 4.2 to 12 or 24 K, the AMR variations decrease significantly in amplitude, and the patterns of variations as a function of bias voltage become smoothed (see Figs. 1 and 2).

To analyze the temperature dependence of the enhanced AMR quantitatively, we compare to the theory of resistance variations due to quantum interference.[15,16] For this purpose,

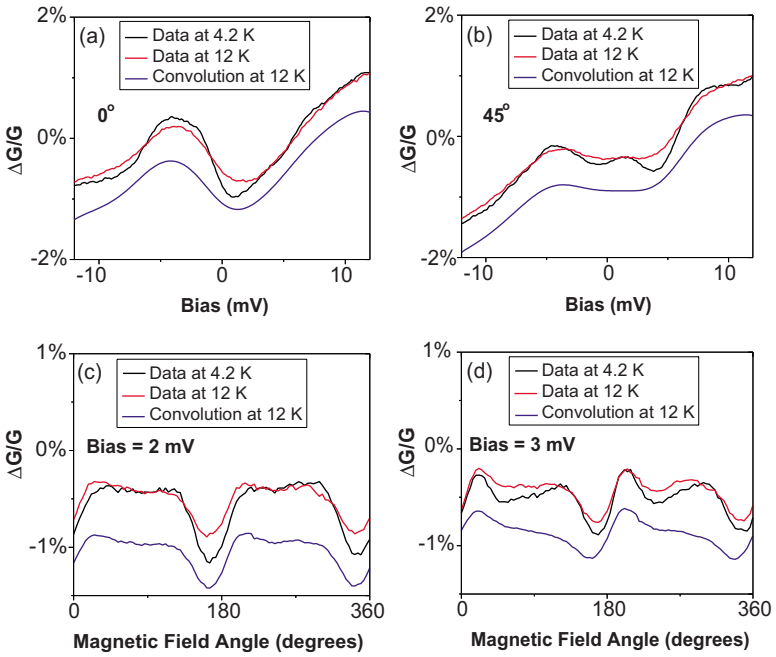

FIG. 3. (Color online) Comparisons between measured conductance (dI/dV) fluctuations at 12 K and the fluctuations predicted from the convolution of the 4.2 K data with the derivative of a Fermi function at 12 K [see Eq. (3)]. [(a) and (b)] dI/dV versus bias at fixed magnetic-field angles of 0° and 45°. [(c) and (d)] dI/dV versus magnetic-field angle at fixed biases of 2 and 3 mV. In all four panels, the curves corresponding to the convolution at 12 K are artificially shifted down by 0.5% for clarity. All data are from the same 4.05 kΩ Permalloy junction shown in Fig. 2.

it is more straightforward to analyze the conductance, dI/dV, rather than the resistance. For the case of a nanoscale contact, in the limit where the resistance of the electrodes adjacent to the constriction is much less than the total contact resistance, the contributions to quantum interference from the two electrodes are expected to separate and become simply additive.[16] The primary mechanism causing the conductance to be temperature dependent should be simple energy averaging of the contributions of electrons over a range on the scale of k_BT. Quantitatively, the current as a function of bias (V) and temperature (T) is predicted to have the form[15,16]

$$I(V,T) \propto \frac{e}{h}\int \{1 + g_L[\varepsilon - \mu_L(V)] + g_R[\varepsilon - \mu_R(V)]\} \times \{f_F[\varepsilon - \mu_L(V)] - f_F[\varepsilon - \mu_R(V)]\}d\varepsilon, \quad (1)$$

where g_L and g_R describe the energy-dependent contributions to the device transmission from quantum interference in the left and right electrodes, f_F is the Fermi function, $\mu_L(V)$ and $\mu_R(V)$ are the chemical potentials in the left and right electrodes with $\mu_L(V) - \mu_R(V) = eV$, and energies are measured relative to the Fermi energy. After separating the integrals, changing variables of integration, and taking the derivative with respect to voltage, we have for the conductance

$$\frac{dI}{dV}(V,T) \propto \text{const} - \frac{e^2}{h}\int \left[g_L(\varepsilon)\frac{df_F}{d\varepsilon}(\varepsilon + eV) + g_R(\varepsilon)\frac{df_F}{d\varepsilon}(\varepsilon - eV)\right]d\varepsilon. \quad (2)$$

This implies that $dI/dV(V,T)$ at nonzero temperature can be determined by convolving the zero-temperature conductance with the derivative of a Fermi function,

$$\frac{dI}{dV}(V,T) = -\int \frac{dI}{dV}(\varepsilon/e, T=0)\frac{df_F}{d\varepsilon}(eV - \varepsilon)d\varepsilon. \quad (3)$$

To compare the prediction of Eq. (3) to the experimental data, we approximate the zero-temperature conductance by $dI/dV(V)$ measured at 4.2 K and convolve with the derivative of the Fermi function at the experimental temperature. The results are plotted in Fig. 3, for the same Py sample shown in Fig. 2. Figures 3(a) and 3(b) show the bias dependence of the conductance at 4.2 and 12 K at two angles of applied magnetic field. The convolutions with a Fermi function reproduce the 12 K data very well, with no adjustable fitting parameters. Figures 3(c) and 3(d) show the dependence of conductance on the angle of the magnetic field at 4.2 and 12 K for the same sample at two different values of bias. To compare to theory for these curves, we convolve the bias dependence of the conductance with the derivative of the Fermi function at each value of field angle, and then plot the angular dependence of the result at the selected bias points. Again, the agreement is excellent, with no free parameters. A similar quality of agreement is seen at all of the values of bias and angle we have explored, for four samples (two Py, one Co, and one Ni). We do not attempt to make a quantitative comparison to the 24 K data in Fig. 2(c) because we did not collect the 4.2 K data over a sufficiently wide bias range for an accurate convolution at 24 K, but we see no features inconsistent with simple energy averaging.

The fact that we can express the temperature dependence of the enhanced AMR by a simple convolution of the measured low-temperature bias dependence with the derivative of a Fermi function is consistent with the proposal that the AMR is produced by quantum interference in the contact,[3,5] but this is not the only physical mechanism consistent with our result. Other mechanisms that result in an energy-dependent transmission factor would produce a temperature dependence governed by energy averaging, similar to that

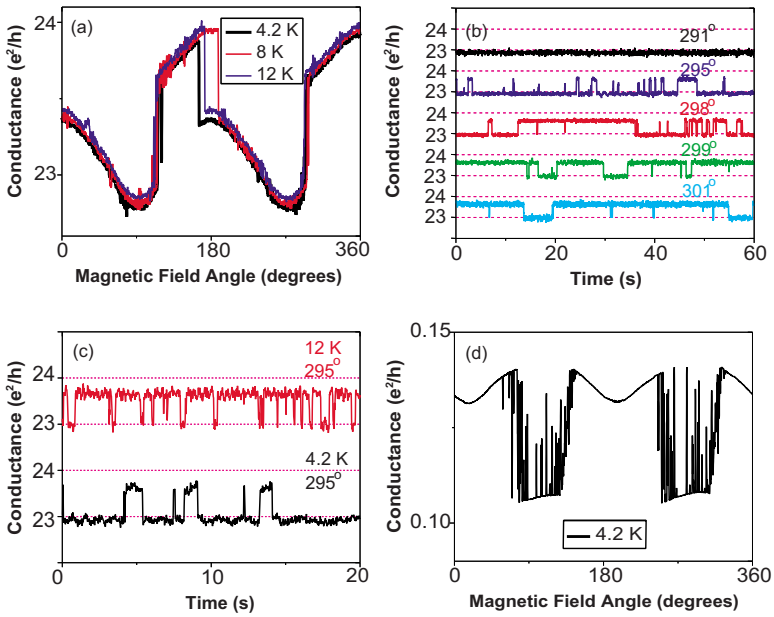

FIG. 4. (Color online) [(a) and (b)] Abrupt changes in conductance for a Ni junction with an average resistance of 1.1 kΩ (a) as a function of magnetic-field angle at three temperatures and (b) as a function of time at 4.2 K. (c) Time-dependent conductance fluctuations for the same 1.1 kΩ Ni junction at temperatures of 4.2 and 12 K. (d) Fluctuations in conductance as a function of magnetic-field angle for a Permalloy junction in the tunneling regime (average resistance of 170 kΩ) at 4.2 K. In (a) and (d), the sweep speed is 12 deg/minute.

described by Eq. (3). For example, transport via resonant states in tunnel junctions[17] would have a similar form of temperature dependence, although we do not believe that this model should apply for the devices we discuss here because they have direct metallic transport. However, our measurements do provide evidence to rule out alternative mechanisms that might produce changes in conductance as a function of bias and field angle on account of structural changes in the junction, for instance, due to magnetostriction or magnetostatic forces,[18,19] because we would not expect these to have this simple form of temperature dependence.

III. FIELD-ANGLE DEPENDENT ATOMIC REARRANGEMENTS

While we do not believe that structural instabilities can explain the smoothly varying enhanced AMR signals that we observed at low temperature, we find that structural instabilities do become increasingly important as the temperature is increased. All of the ferromagnetic contacts we have studied exhibit thermally activated time-dependent resistance noise and irreversible changes in resistance at temperatures above a few tens of Kelvin. A few samples (~10%) show time-dependent switching and other large transport artifacts even at 4.2 K at particular values of magnetic-field angle, as we discuss below. In general, we find that as the temperature is first increased from 4.2 K, the time-dependent signals initially take the form of simple "telegraph-type" two-level fluctuations, which switch back and forth between two resistance values. With further increases in temperature, the switching rates increase, so that these initial fluctuations may become faster than can be measured in the experimental bandwidth, while a large number of additional fluctuators become active. At temperatures around 60 K, a sufficiently large number of fluctuators are active that irreversible changes in the contact resistance become common on time scales of 10 min. Observations similar to these have been made previously in nanometer-scale contacts formed from a wide variety of nonferromagnetic metals, and the fluctuations have been identified as due to thermally activated transitions of atoms or small groups of atoms between different configurations in the contact region, with switching rates governed by a wide distribution of activation energies.[7–9] From our measurements, we conclude that nanoscale metal contacts made by the electromigration process using Permalloy, Ni, or Co are generally highly dynamic structures at temperatures above 100–150 K, with atoms undergoing transitions between different positions on time scales much faster than the typical scales of transport experiments.

We attribute one particularly striking type of experimental observation to structural instabilities. Figure 4(a) shows measurements of conductance versus the angle of an applied magnetic field for a Ni device at low temperature. Near particular values of magnetic-field angle that are reproducible for a given sample, the conductance undergoes sharp jumps with magnitude on the order of e^2/h. These data resemble a prediction[10] that nanoscale ferromagnetic contacts should exhibit sharp conductance steps versus field angle due to BAMR. This prediction has been cited to explain recent experiments on Fe and Co contacts formed by different procedures than electromigration.[4,11] However, measurements of the time dependence of the conductance in our devices near the switching points indicate that the transitions we observed are due to atomic reconfigurations rather than an intrinsic electronic effect such as BAMR. If we set the magnetic field at an angle close to the transition region, we observe two-level fluctuations as a function of time, with magnitude equal to the conductance change at the step. The switching rates of the fluctuator depend very sensitively on the field direction [Fig. 4(b)] (see also similar data from a different sample in our previous comment[12]). Over a range of field angle of about 10°, the dynamics changes gradually from a situation in which the sample is in the high conductance state almost 100% of the time, through switching with a duty cycle that is approximately 50% in each state, to being in the low con-

ductance state 100% of the time. This indicates that the relative energy of the two metastable atomic configurations depends very sensitively on the field angle, and the abrupt conductance steps observed in Fig. 4(a) happen because the atomic ground state switches from one configuration to the other as the field angle is changed. The switching rates become faster as a function of increased temperature [Fig. 4(c)] as anticipated for thermally activated transitions. We have considered the possibility that the time-dependent two-level fluctuations might simply be due to a background fluctuator present in the sample over a broad range of field angle and unrelated to the conductance step, but whose amplitude might be visible only near the conductance step because its conductance change might be amplified by the BAMR effect. We can rule out this possibility because the change in the duty cycle of the fluctuator over a narrow range of field angle about the position of the conductance step indicates that the conductance step and the two-level fluctuator are both caused by the motion of the same atom or small group of atoms.

Structural instabilities and atomic motion can also sometimes cause more complicated artifacts than the abrupt two-level switching shown in Fig. 4(a). For example, Fig. 4(d) shows a trace of conductance versus magnetic-field angle for a Permalloy contact in the tunneling regime (with a conductance less than e^2/h), which for a majority of the angular range shows a smooth evolution of conductance but for reproducible segments of angle switches to a different state with both a lower average conductance and fast switching between two conductance states.

Whenever we have observed abrupt changes in conductance as a function of magnetic-field angle, like those shown in Fig. 4, we have also observed time-dependent two-level switching for field values near the conductance step. From this, we do *not* conclude that the theory of BAMR is incorrect. However, our measurements do show that the atomic configuration within ferromagnetic metal contacts can change as a function of the angle of an applied magnetic field, producing abrupt steps in conductance at field angles that are reproducible in a given sample. Because of this sensitivity, special care will be required in order to test conclusively whether or not BAMR exists as predicted.

IV. CONCLUSIONS

We have studied the temperature dependence of electron transport in nanoscale ferromagnetic contacts made using electromigration. We find that the enhanced AMR signals observed previously[3] have the temperature dependence expected for signals due to quantum interference of electrons near the contact region. The amplitude of the enhanced AMR signals decreases quickly as the temperature is raised because their bias dependence smoothes out due to energy averaging and, as a consequence, the enhanced AMR signals are no longer significant well above cryogenic temperatures. The other effect of temperature that we have observed in these samples is that time-dependent changes in atomic configuration are present within all devices above a few tens of Kelvin. In a small fraction of devices ($\sim$10%), time-dependent two-level fluctuations can be observed even at 4.2 K. These instabilities can be very sensitive to the angle of an applied magnetic field, leading to abrupt steps in conductance that mimic the signals expected for the BAMR effect.

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