Supporting Online Material for

Measurement of Fast Electron Spin Relaxation Times with Atomic Resolution

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Movie S1

Supporting Online Material:
Measurement of fast electron spin relaxation times with atomic resolution

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Experimental Setup:

We use an ultra-high-vacuum scanning tunneling microscope (STM) operating at an adjustable temperature of 0.6 K to 10 K. Magnetic fields up to 7 T were applied perpendicular to the sample surface. The fast voltage pulses are applied to the STM tip, but throughout this report we use the commonly employed convention of specifying the voltage of the sample with respect to the tip. For pump-probe measurements the tip-sample distance was fixed by setting the tunnel current to 1 nA at +10.0 mV sample voltage prior to opening the feedback loop.

The pump and probe voltage pulses were generated as continous pulse trains by a pulse pattern generator (Agilent 81110A). The pump-probe cycle was repeated every 2 μs and the probe pulse was chopped at 810 Hz. The pump and probe pulses were summed, attenuated by 20 dB and applied to the tip of the STM. The tunnel current is detected at the sample and fed to a current preamplifier (Femto DLPCA-200) with ~1 kHz bandwidth and from there to a lockin amplifier to selectively detect the 810 Hz component of the current corresponding to the tunnel current of the probe pulse. For the measurements shown in Fig. 2C, Fig. 4 and Fig. S2 the pump and probe pulses were 100 ns FWHM long with 50 ns linear-ramp rise and fall times. The amplitude of the pump pulse was −36.5 mV and thereby well above the −16.7 mV threshold for spin excitation. The probe pulse voltage was −4.0 mV leading to a baseline of N = 341 electrons per probe pulse as measured when the probe precedes the pump. The pulse parameters used in Fig. 3A are the same except for a lowered probe-pulse voltage of −1.2 mV and the variable pump-pulse voltage.

Spin-polarized STM measurements on Fe-Cu dimers:

The spin-polarized tip used in this work has one magnetic atom, Mn, attached to the otherwise non-magnetic Cu-coated apex. The magnetic atom that was used to create spin-polarization in the tip was picked up after the Fe-Cu dimers were assembled by vertical atom manipulation *(S1, S2)*. The magnetic moment of the attached atom is aligned parallel to the external magnetic field. This also determines the direction of the tip's spin-polarization *(S3)*. Since the magnetic atom at the tip apex is adsorbed directly on the metal surface of the tip (without a decoupling layer such as Cu_2N) much shorter spin lifetimes can be expected. Indeed we did not observe spin excitations for the tip atom and detect no dynamical change in the tip's spin polarization for all timescales that we can access experimentally.

In the present experiments, the magnetic field was applied perpendicular to the sample surface. Hence the tip's direction of spin polarization is also perpendicular to the sample surface. Since we see a strong signal in the spin sensitive pump-probe measurements of the Fe-Cu dimer (Fig. 2C) we conclude that the direction of its easy axis has a significant component parallel to the tip's spin polarization and therefore perpendicular to the sample surface. On the other hand the magnetic field dependence of the spin relaxation time (Fig. 4) indicates that magnetic field and easy-axis anisotropy are not completely parallel.
We note that a single Fe atom on the Cu binding site on Cu_2N has an easy axis that lies in the plane of the sample *(S4)*. In contrast to the single Fe atom, the anisotropy axis of the Fe-Cu dimer is rotated largely out of the sample plane. However, within the present data set we can not quantitatively determine the direction of the easy-axis of the Fe-Cu dimer.

Cross correlation of Pump and Probe pulse:

The linear slope in the differential conductance of the Fe-Cu dimer (see Fig. 2B) gives a quadratic (non-linear) shape to the $I(V)$-curve of the tunnel junction *(S5)*. Since the voltages for pump and probe pulse are summed, the voltage across the tunnel junction increases beyond V_{pump} or V_{probe} when pump and probe overlap. Hence the non-linearity of the tunnel junction creates a component of the current which is proportional to the cross correlation of the pump and probe voltages when the pulses overlap in time . This signal can be used to monitor the quality of the pulses directly at the tunnel junction (Fig. S1). Note that this cross correlation signal is present in all three panels of Fig. 2C which confirms that it is not of magnetic origin.

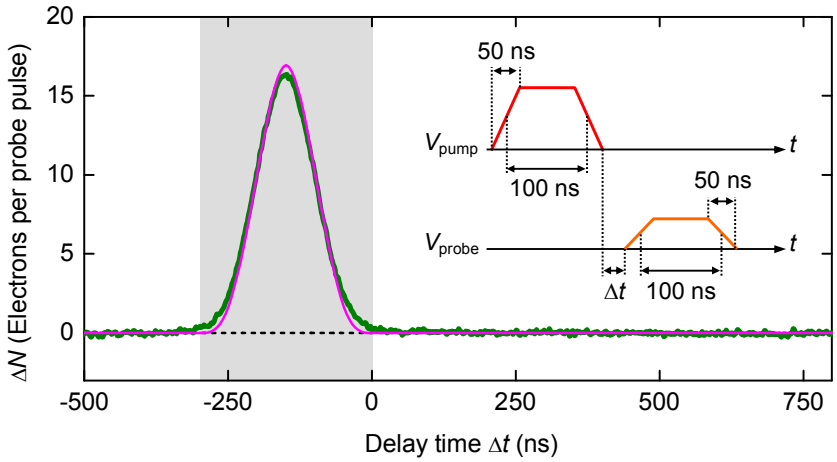

Figure S1. Pump-probe cross correlation. Green curve: ΔN vs. Δt of an Fe-Cu dimer, measured with the pump voltage well below the threshold for creating spin excitations. The gray region corresponds to the delay times in which overlap between the pump and probe pulses occurs assuming the ideal pulse shapes (see inset). A cross correlation term due to a non-linearity in the current vs. voltage characteristics of the tunnel junction (see Fig. 2B) gives rise to the peak seen in the gray region. Purple curve: Calculated ΔN vs. Δt for the experimentally derived values of pump amplitude $V_{\mathrm{pump}} = -8.7$ mV, probe amplitude $V_{\mathrm{probe}} = -1.2$ mV, average differential conductance of the tunnel junction $\mathrm{d}I/\mathrm{d}V(V = 0\ \mathrm{mV}) = 0.11$ nA/mV, average slope in the differential conductance $\mathrm{d}^2I/\mathrm{d}V^2 = -2.1$ pA/mV^2 and the pulse shapes shown in the inset. The baseline number of electrons per probe pulse is 218.

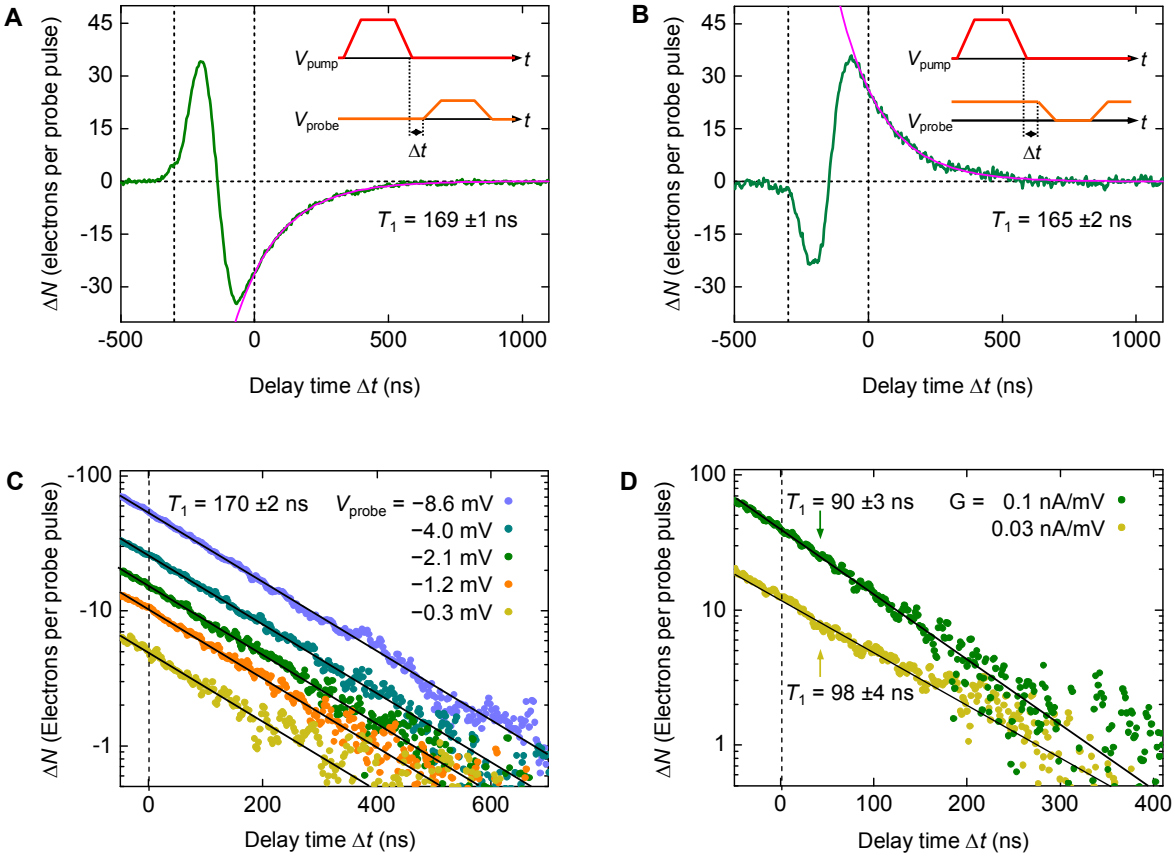

Figure S2. Additional control experiments demonstrating that the tunnel current during the probe pulse has no impact on the spin relaxation time. All measurements were performed on the Fe-Cu dimer represented with green dots in Fig. 4B. (**A**) Pump-probe measurement, ΔN vs. Δt, at $B = 5$ T with the pump and probe pulses having a width of 100 ns at FWHM and 50 ns linear-ramp rise and fall (see inset and above section *Experimental Setup*). The voltage of the probe pulse is $V_{probe} = -4$ mV. (**B**) Pump-probe measurement with the same conditions as in *(A)* but with an inverted probe pulse: A voltage of −4 mV was constantly applied except for a window of 100 ns at FWHM. As expected the measured signal is the inverse of that recorded in *(A)*. This scheme applies a probe pulse that is longer than 1 µs and demonstrates that the Fe-Cu dimer's spin relaxation is not affected by the duration of the probe pulse. (**C**) ΔN vs. Δt at $B = 5$ T with varying probe pulse voltage V_{probe}. The fitted spin relaxation time, T_1, is given by the slope of the black lines in the logarithmic plot. All curves are well fit by a single T_1, so the spin relaxation time is independent of V_{probe}. Together with the pump voltage dependence (Fig. 3B) this demonstrates that the electric field due to the voltage pulses (which is of the order of 10^6 V/cm for the largest voltages used here) does not affect T_1. (**D**) ΔN vs. Δt at $B = 7$ T with varying tip-sample separation identified by the tunnel junction impedance, G, at +10 mV. The exponential fit to the data shows only a slight variation in the measured lifetime that is within the uncertainty of the measurement. This indicates that the magnitude of tunnel current during the pump-probe measurement has no significant impact on the spin relaxation.

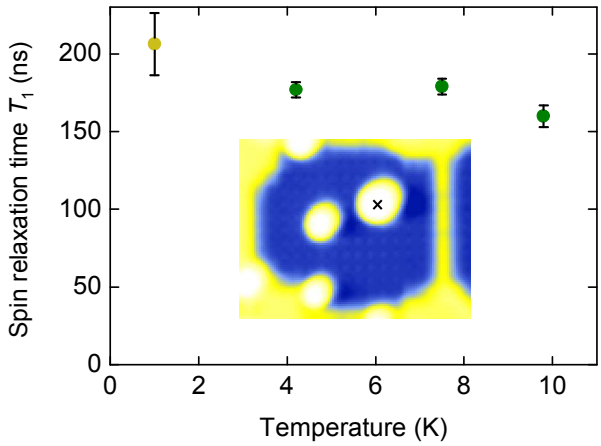

Figure S3. Temperature dependence of the spin relaxation time for an Fe-Cu dimer. For a thermally activated relaxation process a fast drop in T_1 at higher temperatures is expected. However, within the measurement accuracy, T_1 is essentially constant below 10 K. The drop at 10 K might indicate the beginning of a thermally-activated spin relaxation. In either case, at 0.6 K, where all measurements of the main text were recorded, spin relaxation can be attributed to a non-thermal process. The pump-probe measurements >1 K were recorded at 5.5 T magnetic field (green dots). The measurement at 1 K was at 4.0 T magnetic field and was extrapolated to 5.5 T using the measured magnetic field dependence of T_1 which resulted in a bigger uncertainty. Inset: 6.5 nm × 5.0 nm topograph of the Fe-Cu dimer.

References:

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Caption for Supplemental Movie S1, manuscript No. 1191688:

Movie S1. Spatially resolved pump-probe measurement of four Fe-Cu dimers. **(Scene 1)** STM topograph of Fe-Cu dimers that were individually assembled on four Cu_2N patches (green: high; blue: low). Image size is 12 nm by 12 nm. **(Scene 2)** Sequence of pump-probe measurements plotting $\log(|\Delta N|)$ overlaid as color on the topograph for different delay times Δt (red: large signal; white: small signal). The time-dependent signal at $\Delta t > 0$ is localized at each Fe-Cu dimer and decays for increasing Δt. Schematic at left indicates the pulse sequence for each Δt. **(Scene 3)** Comparison of spatially resolved pump-probe measurements for the same Fe-Cu dimers at 4 T and 1 T magnetic field. **(Scene 4)** Spin relaxation times, T_1, for each Fe-Cu dimer as determined by exponential fit to the pump-probe data of (scene 2) and (scene 3). The variations in the T_1 times are likely due to variations in the nearby surface features.

Parameters of the pump-probe measurement: $V_{pump} = -35$ mV, $V_{probe} = -10$ mV, pulse duration 100 ns for the pump pulse and 60 ns for the probe pulse at FWHM with 10 ns linear rise and fall times, repetition of pump-probe cycle every 1.3μs.