Spin-bearing molecules can be stabilized on surfaces and in junctions with desirable properties, such as a net spin that can be adjusted by external stimuli. Using scanning probes, initial and final spin states can be deduced from topographic or spectroscopic data, but how the system transitions between these states is largely unknown. We address this question by manipulating the total spin of magnetic cobalt hydride complexes on a corrugated boron nitride surface with a hydrogen-functionalized scanning probe tip by simultaneously tracking force and conductance. When the additional hydrogen ligand is brought close to the cobalt monohydride, switching between a correlated $S = 1/2$ Kondo state, where host electrons screen the magnetic moment, and an $S = 1$ state with magnetocrystalline anisotropy is observed. We show that the total spin changes when the system is transferred onto a new potential energy surface that is defined by the position of the hydrogen in the junction. These results show how and why chemically functionalized tips are an effective tool to manipulate adatoms and molecules and a promising new method to selectively tune spin systems.

**RESULTS**

Figure 1A shows a constant current image of CoH complexes on h-BN/Rh(111). The lattice mismatch between the Rh(111) substrate and the single monolayer of h-BN results in a strongly corrugated surface with 3.2-nm periodicity, on which the CoH complexes appear as bright protrusions. A clear indication of hydrogen adsorption on the tip apex is the sharp change in tip height, reduced by 20 pm (Fig. 1A, red dashes), while imaging the h-BN/Rh(111) surface in constant current mode (21). Figure 1B shows constant current images of an individual CoH complex located near the rim-valley boundary of h-BN/Rh(111) that is imaged with a hydrogen-functionalized tip. At low junction conductance ($G = I_d/V_S = 1.61 \times 10^{-4}$ $G_0$; $G_0 = 77.48$ $\mu$S; the quantum of conductance), corresponding to relatively large tip-sample separations $z$, the increased contrast due to the hydrogen in the junction partially overlaps a CoH complex (Fig. 1B, bottom panel). As $G$ is increased, this boundary region transitions to a noise-speckled circle with a brighter appearance, that is, larger $z$ height, to compensate for an overall increase in the conductance. Given the strong $G$ dependence within this narrow range, these results hint that the observed contrast is not solely due to the local topography but is also due to mechanical and electronic changes in the junction. These images are qualitatively similar to measurements of undercoordinated metal adatoms in the presence of adsorbed hydrogen (22, 23). Because the hydrogen content of the CoH$_x$ complex governs the spin state ($S$), $d/dV$ spectroscopy was performed while varying the setpoint conductance $G$ with the tip positioned over the central region. At the lowest conductance, $G = 6.45 \times 10^{-4}$ $G_0$ (Fig. 1C, bottom curve), the spectra show two symmetric steps around zero bias with increasing differential conductance. These steps originate from the inelastic spin
excretions of a CoH complex with a total spin \( S = 1 \), where mageneto-crystalline anisotropy has removed the 3d-level degeneracy. Increasing \( G \) by decreasing the tip-sample separation \( z \) results in progressively unstable spectra until the emergence of a stable zero bias peak at \( G = 12.9 \times 10^{-4} G_0 \), identified as an \( S = 1/2 \) CoH2 Kondo resonance (5). This transition is fully reversible, and the initial \( S = 1 \) total spin state is restored when the junction conductance is reduced (see fig. S1). We observe a metastable state, when \( G \) is between \( 8 \times 10^{-3} G_0 \) and \( 11 \times 10^{-3} G_0 \), where the hydride complex randomly transitions between the \( S = 1 \) and \( S = 1/2 \) states on a time scale of 100 ms. The change in tip-sample separation for this conductance range corresponds to a \( \Delta z \) of less than 25 pm. Note that this metastable behavior does not depend on the bias voltage during the spectroscopic measurement (see also fig. S2). Differential conductance \( (dI/dV) \) spectroscopy not only identifies the spin state but also aids in the interpretation of the STM images in Fig. 1B. The constant current images in Fig. 1B were obtained over a bias range (0.3 to 1.6 mV) where the topographic appearance is closely linked to the features in the \( dI/dV \) measurements and, therefore, at small bias voltages, is dominated by the Kondo resonance.

To investigate the switching behavior in detail, we performed \( G(z) \) measurements over CoH complexes and bare h-BN. Approaching h-BN as well as CoH complexes with a bare tip reveals a strictly exponential increase in conductance, \( G(z) = G_0 \exp(-2\kappa z(\sigma_0 + z)) \), where \( \kappa \) is the decay rate and \( \sigma_0 \) is the tip height at the initial setpoint conductance \( G \) (Fig. 2A). Functionalizing the tip apex with hydrogen alters the junction conductance characteristics, with \( G(z) \) showing a less than exponential increase and a reduced \( \kappa \) compared to the data obtained with a bare tip (Fig. 2B). This characteristic behavior is similar to the observations of Weiss et al. (18) on a complex organic molecule with a hydrogen-functionalized tip. However, when approaching a CoH complex with a hydrogen-functionalized tip, \( G(z) \) closely follows the h-BN trace until the conductance rapidly decreases by a factor of 2.5, indicating the \( S = 1 \) to \( S = 1/2 \) transition (Fig. 2B, red) that is observed by local spectroscopy in Fig. 1C. Therefore, the drop in conductance stems from both the direct transfer of hydrogen within the junction, altering the geometry and modifying the tunnel barrier, and the relative change in total conductance between the CoH and CoH2 complexes.

To understand the electronic structure modification in the spin-switching process, we performed DFT calculations for various representative junction geometries (Fig. 2C). A CoH complex on h-BN exhibits a nearly free atom–like electronic structure (Fig. 2C, i), with Stoner split 3d levels giving it a magnetic moment of 2.0 \( \mu_B \) (Bohr magneton) (5). Approaching the complex with a Pt tip, which has a hydrogen atom bound to its apex (Fig. 2C, ii), gradually introduces indirect bonding via the H of the CoH complex, resulting in the reduction of the Co magnetic moment (Fig. 2C, ii). The magnetic moment of Co remains close to 2 \( \mu_B \), an effective \( S = 1 \) state, until the approaching functionalized tip brings the H sp orbitals into direct overlap with the Co d orbitals (Fig. 2C, iii). Bonding of two H atoms to Co, as shown in our previous work, is strong enough to partially quench the magnetic moment of Co, reducing it to 1.2 \( \mu_B \), an effective \( S = 1/2 \) state.

To reveal the microscopic forces at work in the spin transition, we track the frequency shift, \( \Delta f \), of the oscillating tuning fork from its non-interacting resonance frequency, \( f_0 = 29,077 \) Hz. We measure \( \Delta f(z) \) curves...
over switching complexes and the bare h-BN. To remove the long-range forces between the extended tip and the sample, we subtract the background from the data, that is, δF = ΔF_{CoH} − ΔF_{h-BN} (see Materials and Methods and fig. S3). The δF is small and negative before rapidly decreasing upon approach (Fig. 3A, black). This sharp drop in δF coincides with a change in the G(z) measurement similar to that in Fig. 2B; however, here, this feature is broadened because of averaging over the 100-pm oscillation amplitude (Fig. 3A, gray). Short-range forces, F′(z′), were quantified by converting ΔF(z) using the method of Sader and Jarvis (24).

Before the S = 1 to S = 1/2 transition, the force between the tip and sample is weakly attractive and grows exponentially upon approach. As the hydrogen on the tip apex couples to the CoH complex, the attractive force grows steeply over a transition region of 35 pm before leveling off (Fig. 3B). The instantaneous junction conductance, G(z′), is deconvoluted to remove the influence of an oscillating tip (25), revealing that the force and conductance transition regions coincide.

From the F(z′) measurement, we reconstruct the one-dimensional (1D) potential energy landscape, U(z′), across the spin transition by integrating F(z′) (Fig. 3C). The U(z′) curve shows a steep change in slope as the tip brings the hydrogen closer to the CoH complex, suggestive of a transition between potential energy surfaces. This interpretation is broadly in line with a framework recently developed by Hapala et al. (26) to describe high-resolution AFM imaging with functionalized tips.

One key component of this model is that the probe particle, hydrogen in our case, not only follows the lowest potential energy surface but also undergoes relaxation within the junction. In our U(z′) measurement, the kink corresponds to the relative z′ distance where the hydrogen on the tip apex can relax onto a lower potential energy surface. Chemically, CoH is transformed into CoH2, a magnetically distinct complex with a different potential energy surface. DFT calculations show that the transition between the two distinct chemical configurations proceeds via a continuum of intermediate transition states. This intermediate regime is characterized by hydrogen on the tip sharing its bond with the tip apex and the CoH while simultaneously undergoing reorientation within the junction. In tackling this complexity, it is instructive to examine the diabatic potential energy surfaces in the limiting cases, that is, when CoH interacts with a hydrogen-functionalized tip and when CoH2 interacts with a clean metallic tip. In Fig. 3D, we plot the dependence of the total energies of the two abovementioned configurations (red dash-dotted and blue dashed curves, respectively) on the

![Fig. 3. Force measurements on a switching complex.](http://advances.sciencemag.org/)
tip-sample distance \(d\) (see Materials and Methods and fig. S4), the chosen reaction coordinate for our system. The intersection of the two potential energy curves with varying junction width confirms that hydrogen transfer drives the spin transition and is consistent with experimental observations. As mentioned above, the system does not follow the diabatic curves; rather, it undergoes a barrierless transition along a reaction path difficult to precisely identify by ab initio methods because of the shallowness and unevenness of the hydrogen potential energy landscape in the junction. An approximate shape of this pathway is shown in Fig. 3D, with a gray dotted line linking the two asymptotic cases. The presence of a transition region (gray region in Fig. 3, B and C) further indicates that the spin switching is driven by the approaching hydrogen and that the residence time is shorter than the measurement time, leading to averaging and smearing-out when probed with the oscillating tip. Furthermore, the increased mechanical dissipation during the transition (~55 meV per cycle) (see fig. S5) is in good agreement with the potential energy difference across the transition region of the \(U(z')\) measurement (~35 meV) and points to the existence of hysteresis on the time scale of the oscillation period of \((2f_0)^{-1} \sim 15\) \(\mu s\).

**DISCUSSION**

We have reversibly switched the spin state of cobalt hydride complexes in a tunnel junction by using the hydrogen on the tip apex as a tunable ligand. During the total spin change, a transition region exists where the hydrogen can occupy two nearly equivalent sites separated only by a negligible barrier. As the junction conductance is altered, site equivalence is removed, and the tip displacement shifts the system to a preferred site. Beyond total spin, interactions with the tip determine how the cobalt spin couples with the host electron bath and whether a correlated Kondo state emerges. The relative stability of the double potential well and the possible occurrence of correlations are expected to be dependent on the materials used and the reactivity of the adsorbed molecule on the tip and the undercoordinated adatom. Reconstructing the 1D potential energy landscape shows that chemical transformation within the junction is responsible for the change of the total spin. Our measurements highlight how tip functionalization can influence the spin under investigation and suggests that they may be used to tailor molecular spins in ways difficult to achieve through traditional chemical synthesis.

**MATERIALS AND METHODS**

The clean Rh(111) surface was prepared by multiple cycles of argon ion sputtering and annealing to 1100 K in an ultrahigh vacuum chamber. During the final annealing cycle, borazine (\(\text{B}_3\text{H}_6\)) gas was exposed to the hot sample at a pressure of \(1.2 \times 10^{-6}\) mbar for about ~2 min, resulting in a monatomic layer of \(h\)-BN. Subsequently, the sample was cooled down (~20 K), and cobalt was deposited via an electron beam evaporator. Because hydrogen is the predominant component of the residual gas background, it is responsible for the formation of the cobalt hydride complexes. STM/AFM experiments were performed on a homebuilt instrument operating in ultrahigh vacuum at a base temperature of 1.1 K. All spectroscopic \((dI/dV)\) measurements were obtained by adding a small sinusoidal voltage \((V_{\text{mod}} = 0.2 \text{ mV}, f_{\text{mod}} = 600 \text{ to } 800 \text{ Hz})\) to the bias voltage \(V\) and using an external lock-in amplifier. We used \(V' = -10 \text{ mV}\) to adjust the conductance \(G\) before recording \(dI/dV\) curves by varying \(V\) at constant \(z\), as well as to record \(G(z)\) and \(\Delta f(z)\) curves at constant \(V\). Hydrogen pickup occurred fortuitously as the experiments went on, for example, during the approach of an initial CoH\(_x\) \((x = 1 \text{ to } 2)\) complex with a nonfunctionalized tip. Hydrogen-terminated tips were initially identified by features such as enhanced contrast in constant current imaging and then verified by the characteristic nonexponential behavior in \(G(z)\) measurements (Fig. 2B, green).

The quartz tuning fork has a resonance frequency of \(f_0 = 29,077\) Hz and a quality factor of approximately 10,000. Oscillation amplitudes of 100 pm were used. For the frequency shift curves, \(\Delta f(z)\), the tip was positioned above the CoH complex with the oscillating tuning fork. The feedback loop was disabled, and the oscillating tip then approached 200 to 300 pm toward the sample and back. Immediately after completion, a retract curve was obtained by moving 2000 pm away from the surface (see fig. S3). When this sequence was completed, the tip was moved at constant height to the bare \(h\)-BN, and \(\Delta f(BN)\) was obtained. The frequency shift due to short-range forces was obtained by taking the difference \(\Delta f = \Delta f_{\text{CoH}} - \Delta f_{\text{BN}}\). We distinguish between time-averaged signals at tip heights \(z\) and deconvoluted (instantaneous) signals at tip heights \(z'\).

First-principles calculations were carried out in a manner consistent with the calculations used in our previous work (5). Briefly, a DFT approach, based on the projector augmented wave method (27) and a plane wave basis set (28), was used as implemented in the Vienna Ab initio Simulation Package (29). Exchange and correlation were treated with the gradient-corrected functional as formalized in PBE (Perdew, Burke, Ernzerhof) (30). On-site Coulomb interaction corrections were accounted for in the framework of the LSDA+U (local spin density approximation including Hubbard U) formalism, as introduced by Dudarev et al. (31). The considered geometry of the system was identical to the one introduced in the study by Jacobson et al. (5), that is, a Rh(111) surface was simulated by five Rh layers, on top of which an \(h\)-BN sheet was deposited (considered to be commensurate for computational feasibility reasons). A CoH complex was residing on top of an N atom of the \(h\)-BN sheet. The tip was simulated by a Pt pyramid of four atoms attached to an Rh surface. Before the tip approach simulation, the tip and the sample were allowed to relax. After placing the tip in the vicinity of the CoH\(_x\) complex, the CoH\(_x\) complex and the apex atom of the tip (as well as the attached hydrogen atom, where appropriate) were allowed to relax again, assuming an equilibrium static configuration. A full relaxation of the tip and the \(h\)-BN atoms around the CoH\(_x\) complex was tested and not found to introduce any change to the results above the calculation error level. In the experimentally relevant case of a CoH complex being approached with a singly hydrogenated tip, we are thus left with four atoms allowed to relax in three dimensions, amounting to a 12D configuration space. However, in effect, both the apex Pt and the Co atoms were found to experience only vertical relaxations, which, according to our tests, result in a minor quantitative change of the total energies and magnetic moments. This complies with our understanding of the mechanism of CoH\(_x\) complex spin switching, the latter being governed by the Co–H bonding, which, in turn, is altered through the rearrangement of the hydrogen atoms. To underline this fact, the calculation results presented in Fig. 3D and fig. S6 were obtained in a diabatic fashion, meaning that the initial relative orientation of the CoH\(_x\) and the TipH\(_{3-x}\) atoms were kept frozen and that only the distance between the tip and the sample (characterized by the distance \(d\) between Co and apex Pt atoms) (see fig. S4) was varied. For hydrogen adsorption and transfer energies as well as the adiabatic transition calculation, the junction was allowed to relax.

**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/3/4/e1602060/DC1

fig. S1. Reversibility of the switching process.
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