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Electromigration of single metal atoms observed by scanning tunneling microscopy

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The authors show in this letter that single metal atoms on a Ni(111) surface can be pushed by electromigration forces from a scanning tunneling microscope tip. This repulsive interaction is observed over a length scale of 6 nm. While for voltages above −300 mV the atoms are pulled by the microscope tip, the atoms are pushed away below this threshold. This migration is explained by a resonant scattering of strongly correlated electrons. At small voltages chemical forces are pulling the atom, while for larger voltages the atomic manipulation is assisted by the tunneling current. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2430102]

Nanotechnology is rapidly evolving, but the control of nanomaterials at the atomic scale can still be considered as one of the outmost challenges. It has been demonstrated that single atoms can be positioned by a scanning tunneling microscope (STM) tip.[1] This technique has been shown to be unique and can even be used to assemble structures up to several hundreds of atoms. Putting together structures in a one-by-one manner is time consuming and therefore the size and number of structures are limited. Single atoms then again can also be moved by electromigration, which has the potential to move many atoms simultaneously. There a lateral current moves adsorbates and has been effectively used to create smooth and defect-free terraces on semiconductor materials.[2] Step bunching, meandering, and pairing are effects caused by electromigration. Here the atoms react on an effective electric field and a momentum transfer from the current carriers.[3] Recently it was also applied to create electrodes with atomic sized tips for single molecule contacts in break junctions.[4] We show here that single Au atoms can be moved by electromigration forces involving *d* electrons from the surface resonance on the Ni(111) surface. The data shown in this letter have been taken with a STM operating at low temperatures. Experiments for lateral manipulation are taken at different voltages and show that the atoms are either pulled to the microscopes tip or pushed away from it. The observed effects are explained by a chemical interaction between the tip and adatom together with a local heating effect from the current. In the repulsive energy range, current and field effects let the atoms hop into adjacent adsorption sites away from the tip.

The measurements and analysis of the experiment follow the procedure described in detail in Ref. 5. A single crystal Ni(111) was cleaned by repeated Ne^+ ion sputtering and annealing cycles. The cleaning was then followed by dosing of 10 l of oxygen and an annealing period with a temperature above 1300 K. For the measurements with STM, the sample was then cooled by liquid helium before transferring it into the microscope, which was kept at 5 K during the measurements. Large terraces with a defect concentration of less than 0.01% were achieved. Single Au atoms were deposited onto the substrate surface *in situ* at low temperature from a resistively heated evaporator.

Manipulation experiments were performed by moving the tip along a straight path on the surface starting on the atom, as it is shown in Fig. 1. The feedback loop was closed to keep a constant height. The current and voltage settings were then changed over a given interval. The ratio of the atom path and the tip path is a measure of whether the manipulation is working or not. A lateral manipulation was considered successful if the atom moves together with the STM

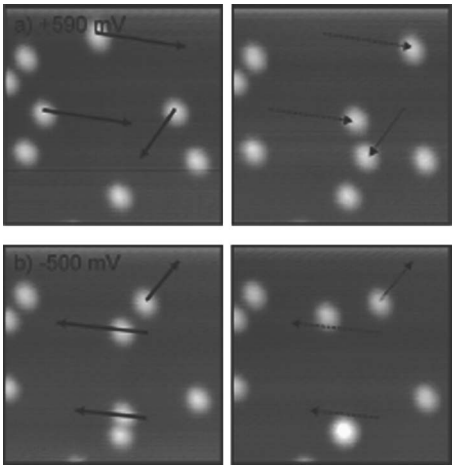

FIG. 1. STM images show lateral manipulation experiment of single Au atoms on a Ni(111) surface. The topography images (80×80 Å^2, $U=-94$ mV, and $I=0.4$ nA) show the single atoms and the black arrows indicate the tip paths at a closer distance to move the atoms ($I=200$ nA). (a) While for all positive voltages the atoms follow the microscope tip, (b) at negative voltage below −400 mV the atoms move away from the tip.

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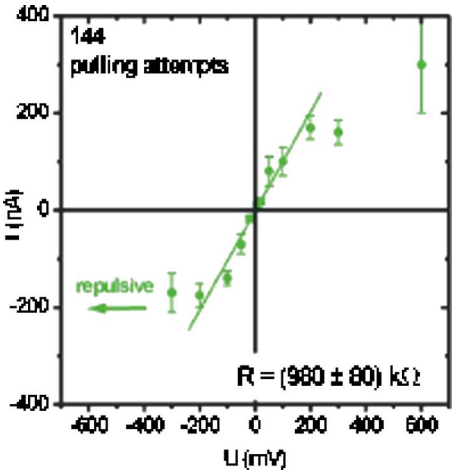

FIG. 2. (Color online) Current threshold as a function of voltage is linear in the interval of ±200 mV. Above ±200 mV the atoms start to move already at a closer distance, while below −300 mV the tip-atom interaction becomes repulsive.

tip, cf. Fig. 1(a). For a fixed voltage the current is increased until the manipulations become successful. The current threshold as a function of voltage is shown in Fig. 2. A linear dependence of the current threshold is observed over the voltage interval of ±200 mV. Over this range of voltages chemical forces are pulling the atom.[5] Both van der Waals and metal adhesion forces are contributing in this range of tip-atom distances.[6] The slope of the current threshold versus the voltage can be expressed as a tunneling resistance threshold of 900 kΩ, a comparatively small value. The Au atoms are easier to move than Ag and Au atoms on a Ag(111) surface, where threshold values of 184 and 130 kΩ have been determined.[5] By using tunneling voltages and currents corresponding to a tunneling resistance above the threshold value, the atoms do not move. The threshold value is a measure of the exerted force from the microscope tip and the migration barrier which needs to be overcome by the atom in order to jump to the next adsorption site. By increasing the voltage above ±200 mV, the atom already starts to move at a smaller current. This effect has been also observed for Au atoms on a Ag(111) substrate and is attributed to a local heating of the atoms bonded to the surface.[5] Surprisingly the atoms are pushed away from the tip if voltages below −400 mV are used, cf. Fig. 1(b).

To investigate this repulsive interaction a second type of experiment has been conducted. In Fig. 3 a quantum corral from 24 Au atoms is shown, which has been constructed by lateral manipulation. By applying a voltage pulse of −600 mV in the center of the corral, the atoms are pushed outwards. A smaller corral constructed from nine Ag atoms is shown in Figs. 3(c)–3(f). The tip is placed in the center of the construction, and again, by applying a pulse with negative voltage the atoms are pushed outwards. Although the hopping is essentially radially outwards, a small amount moves tangentially too. In Figs. 3(a) and 3(b) a fraction of 0.4 of the atoms makes single fcc-hcp or hcp-fcc jumps to the next site, while an amount of 0.2 of the atoms makes fcc-fcc or hcp-hcp jumps with twice the jump length. The Ag atoms in Figs. 2(c)–2(f) move less easily; here the values are 0.2 for single jumps and 0.2 for double jump length. In case of a large number of atoms, double jumps are expected on average to occur less frequently.

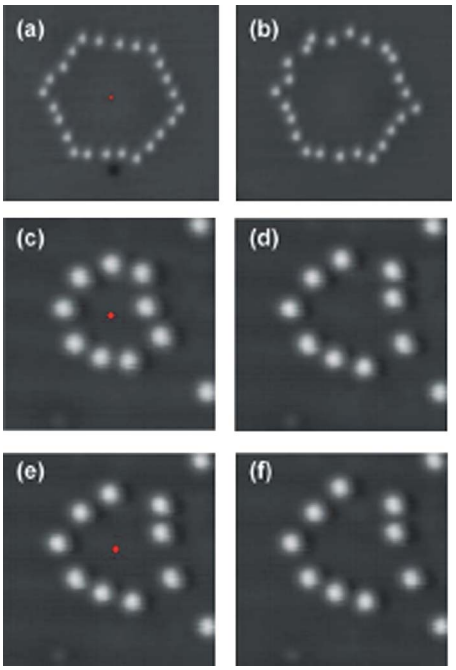

FIG. 3. (a) Image (20×20 nm^2) of 24 Au atoms arranged in a hexagon shift to new adsorption sites if a voltage pulse of −600 meV is applied in the center. (b) The atoms jump outwards although they are 6 nm away from the tip. (c)–(f) The same effect can be observed with Ag atoms (8×8 nm^2) here the maximal range is 3 nm.

In electromigration one considers freelike electrons ballistically with transfer of momentum by scattering with an impurity. The electric field gives another contribution by acting directly on the migrating ion. These forces point in opposing directions and the resulting force depends on the magnitude of each contribution. Upon reversal of the polarity the direction of the resulting force reverses too.[7]

The atoms in the experiment shown in Fig. 3 remain stable for positive voltages, while for negative voltages they are pushed away from the tip. Furthermore, we observe this interaction even at a distance of 6 nm and we can therefore exclude a chemical interaction, which is short ranged. The movement of single atoms induced by a current has been investigated for Cl atoms on a Si(111) surface.[8] There Cl atoms started to migrate by using electrons with an energy of the surface state band at positive voltage. Upon resonantly trapping an electron, the antibonding state becomes occupied and the bond of the Cl atom to the surface becomes less strong. The atom can then hop to the next adsorption site. We note that a direction is not given.

To explain our experimental findings the electronic structure of substrate and adatom needs to be taken into account. The energy threshold observed in our experiment for a hopping agrees well with the energy of a recently investigated surface resonance with *d* character and downward dispersion.[9] This surface resonance has a binding energy of −243 meV and an effective mass of $m^*=0.4$. By crossing the voltage threshold of −300 meV, the sign of the interaction reverses. Above this threshold essentially bulk electrons are extracted, while below the threshold *d* electrons from the surface resonance are used. These *d* electrons are strongly correlated and show an enhanced Coulomb repulsion. Scattering of these electrons with the adatom is an inelastic Coulomb scattering with transfer of energy and momentum.

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Therefore by changing the voltage, an inelastic Coulomb scattering becomes active.

To understand the direction of the atom's motion one needs to take into account the electric field from the microscope tip. Electric field effects are small, with the voltages typically applied in STM.[5] They can be resonantly enhanced though if the electron current is weakening the bond of the adatom to the substrate or multipolar moments are induced. At negative bias the electric field is pointing away from the tip and pushes the atom then outwards by acting on its electric moments.

The motion of the adatoms is not strictly outwards but shows a tangential component too. It is shown that the vicinity of other adatoms alters the force consistent with the multiple backscattering of charge carriers between adatoms. We understand the observed variation of the direction as a result within a multiple scattering of the surface electrons.[3]

In conclusion we have observed a resonant interaction of *d* electrons with Au atoms on a Ni(111) surface. The atoms are pushed away from the microscope tip even at large distances. The resonant energies coincide with a surface resonance. The migration of single metal atoms on a metallic substrate has been investigated and can be used in switching nanodevices and memory devices. Independent of the microscope tip, external electric fields can be used to functionalize this process.

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