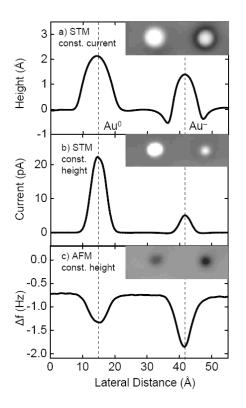
## Individual Atoms and Molecules on Insulating Films Studied with Noncontact AFM

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We investigated the charge state switching of individual gold and silver adatoms on ultrathin NaCl films on Cu(111) using a qPlus tuning fork atomic force microscope (AFM) operated at 5 Kelvin with oscillation amplitudes in the sub-Ångstrom regime. Charging of a gold adatom by one electron charge increased the force on the AFM tip by a few piconewtons. Employing Kelvin probe force microscopy (KPFM) we also measured the local contact potential difference (LCPD) above differently charged adatoms. We observed that the LCPD is shifted depending on the sign of the charge and allows the discrimination of positively charged, neutral, and negatively charged atoms [1].

Furthermore, we modified AFM tips by means of vertical manipulation techniques, i.e. deliberately picking up known adsorbates, to increase spatial resolution. To study the effect of the atomic tip termination we used different well defined tip terminations to image individual pentacene molecules. We compare our experimental results with density functional theory (DFT) calculations to gain insight on the physical origin of contrast formation on the atomic scale.



**Figure 1:** (a) Constant-current scanning tunneling microscopy (STM) measurement (sample bias V = -50 mV, current I = 2 pA) of a neutral (Au<sup>0</sup>, left) and a negatively charged gold adatom (Au<sup>-</sup>, right) adsorbed on NaCl(2ML)/ Cu(111). The line scan is through the center of both adatoms shown in the inset (image size of insets: 55Å × 17Å).

(b) Current (STM measurement) and (c) frequency shift (AFM measurement) recorded simultaneously in constant-height mode (tip height  $\Delta z = 5.0$ Å, V = -5mV, amplitude A = 0.3Å). Due to the increased attractive force above the negatively charged atom the frequency shift is in absolute values larger above Au<sup>-</sup> compared to Au<sup>0</sup>.

[1] L. Gross, F. Mohn, P. Liljeroth, J. Repp, F. J. Giessibl, G. Meyer, Science **2009**, 324, 1428.