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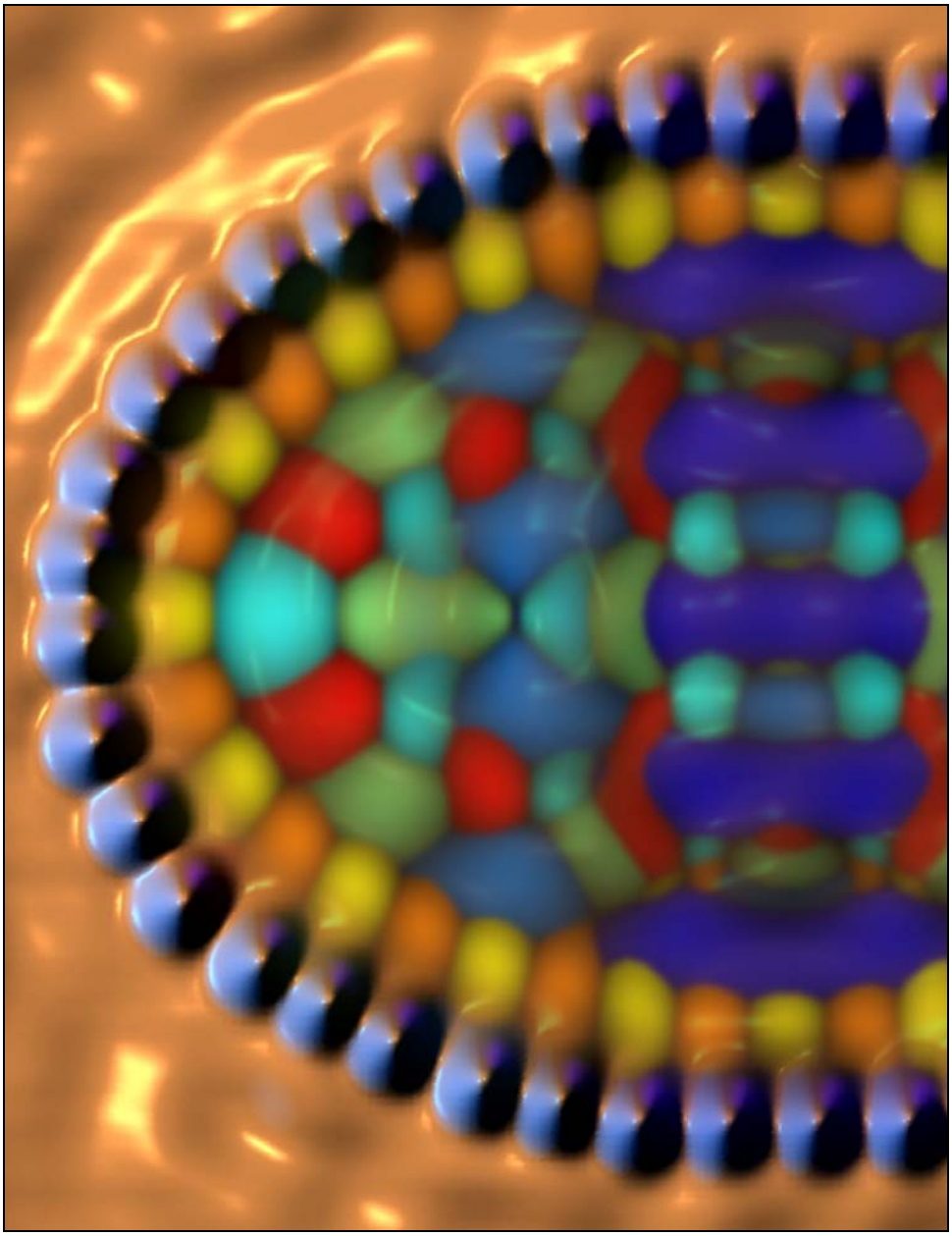
Single-Atom Gating of Quantum State Superpositions

Christopher R. Moon[1], Christopher P. Lutz[2] & Hari C. Manoharan[1*]

[1]*Department of Physics, Stanford University, Stanford, California 94305, USA*

[2]*IBM Almaden Research Center, 650 Harry Road, San Jose, California 95120, USA*

*To whom correspondence should be addressed. E-mail: manoharan@stanford.edu



SIMES, SLAC National Accelerator Center, 2575 Sand Hill Road, Menlo Park, CA 94309

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The ultimate miniaturization of electronic devices will likely require local and coherent control of single electronic wavefunctions. Wavefunctions exist within both physical real space and an abstract state space with a simple geometric interpretation: this state space—or Hilbert space—is spanned by mutually orthogonal state vectors corresponding to the quantized degrees of freedom of the real-space system. Measurement of superpositions is akin to accessing the direction of a vector in Hilbert space, determining an angle of rotation equivalent to quantum phase. Here we show that an individual atom inside a designed quantum corral[1] can control this angle, producing arbitrary coherent superpositions of spatial quantum states. Using scanning tunnelling microscopy and nanostructures assembled atom-by-atom[2] we demonstrate how single spins and quantum mirages[3] can be harnessed to image the superposition of two electronic states. We also present a straightforward method to determine the atom path enacting phase rotations between any desired state vectors. A single atom thus becomes a real-space handle for an abstract Hilbert space, providing a simple technique for coherent quantum state manipulation at the spatial limit of condensed matter.

Progress in quantum nanoscience has engendered a physically diverse array of controllable solid-state quantum systems[4-6]. The prototypical quantum system consists of two wavefunctions that can be coherently combined into superpositions. In this work, we create and study superpositions of electron wavefunctions in nanoassembled quantum corrals where we can finely tune the geometry. These structures permit unique investigations of nanoscale electrons and their correlations, including information propagation[7], lifetime effects[8], Kondo interactions[9], and spin-orbit coupling[10]. We use quantum corrals to take the traditional technique of gating, or the application of electrostatic potentials, to its smallest possible scale. Here, a single adatom couples only to a minute fraction of an electron's spatial extent. Rather than changing applied voltages, we make only geometric changes to the gate position, enabling adiabatic

control of a two-state quantum system. Geometric and adiabatic manipulation of wavefunctions is a robust alternative to dynamic manipulation for various quantum technologies[11, 12].

We engineered an elliptical resonator to harbour degenerate wavefunctions whose superpositions could be manipulated. The solutions to the Schrödinger equation in a hard-walled ellipse possess two quantum numbers: *n*, the number of nodes crossing the minor axis, and *l*, half the number of nodal intersections along the perimeter (these map to the radial and angular momentum quantum numbers in a circle). By judiciously choosing an ellipse's deformation and size, two target wavefunctions can be made degenerate at any energy. We targeted states with even *n* and non-zero *l*, which possess two widely separated regions of concentrated amplitude along the major axis. After analytically solving for the ellipse's eigenspectrum as a function of deformation (Fig. 1a), we aimed to design a corral with states $|n,l\rangle = |4,4\rangle$ and $|2,7\rangle$ degenerate precisely at the Fermi energy E_F. These correspond to the 41st and 42nd most energetic states, or $\psi_j \equiv |j\rangle = |41\rangle$ and $|42\rangle$. On Cu(111), where the surface state band edge is 0.445 eV below E_F and $m^* = 0.38$ bare electron masses[13], this is theoretically achieved in a $2a \times 2b = 157 \times 110$ Å elliptical resonator, whose full energy spectrum is shown in Fig. 1b.

We assembled our designed resonator using a home-built scanning tunnelling microscope (STM) operating in ultrahigh vacuum. The single-crystal Cu(111) substrate was prepared, cooled to ~4 K, and dosed with ~15 Co atoms per (100 Å)^2. We individually manipulated[2] 44 Co adatoms to bound the corral. With spectroscopy, we verified that modes $|41\rangle$ and $|42\rangle$ occurred within a few mV of one another (Supp. Fig. 1). A constant-current (I) topograph of the finished structure is shown in Fig 1c. To confirm that the wavefunctions $|\psi\rangle$ closely describe this system, we used them to calculate (see Methods) a theoretical topograph (Fig. 1d) that reproduces the data

without any fitting parameters. Figure 1e displays the calculated contributions c_j of the significant modes composing the topograph $z(\mathbf{r})$, such that $z(\mathbf{r}) \propto \sum_j c_j \left| \psi_j(\mathbf{r}) \right\rangle \right|^2$.

Next, we added a nanoscopic gate: a single cobalt atom. While moving the adatom across the ellipse—effectively sweeping a local electrostatic potential across the eigenstates—we measured topographs (Fig. 2, first column) and simultaneously acquired dI/dV image maps. By subtracting the dI/dV map of the empty ellipse, we created dI/dV difference maps (Fig. 2, second column). We began by placing the gate atom at one of the maxima of the calculated $|2,7\rangle$ state. The resultant difference map (Fig. 2e) strongly resembles the $|2,7\rangle$ state. Surprisingly, however, when the Co atom was moved rightward to one of the strong maxima of the state $|4,4\rangle$, the image produced (Fig. 2g) was manifestly different from either of the two eigenstates.

We will show that our $\Delta dI/dV$ maps are images of superpositions: phase-coherent $\left| \sum_j a_j |\psi_j\rangle \right|^2$. This is in contrast to typical STM measurements, such as the topograph above, where the sum of tunnelling through independent channels[14] yields signals proportional to phase-insensitive $\sum_j c_j \left| \psi_j \right\rangle \right|^2$. To demonstrate this result, we reproduce the difference maps as linear combinations of the states $|\psi\rangle$ of the unperturbed elliptical corral. These coherent superpositions (Fig. 2, third column) are an excellent match to the mirage data. Any methods neglecting phase interference cannot reproduce our observations (see Supp. Fig. 2).

Electrons in quantum corrals are well modelled by particle-in-a-box solutions to the Schrödinger equation because the surface state wavelength [30 Å in Cu(111) at E_F] is much larger than the spacing between the wall atoms[1, 3, 10, 15-18]. As a first clue to the underlying physics, the original report of the quantum mirage[3] pointed out the similarity between the solitary eigenfunction closest to E_F and the spatial fine structure around the

projected Kondo image. A complementary approach[10, 19-24] treats the wall atoms as discrete scatterers of electron waves and solves the quantum multiple-scattering problem. In fact, the scattering theory was also applied[20] to produce very compelling computed matches to the mirage data and extract the Kondo phase shift. In this prior experimental and theoretical work, the structures studied had modes separated in energy by more than their linewidths—i.e. the state manifold was essentially non-degenerate. Here we focus on the engineered degenerate case where two or more states are forced to coherently superpose by the added gate atom.

While the exact origin of the Kondo resonance in our system is still debated[25, 26], microscopic calculations have indicated that the many-body complexity is manifested in the energy dependence of the density of states while its spatial dependence can be understood from a single-particle perspective[16, 18]. Regardless of its origin, we have verified that Kondo scattering calculations[21] can reproduce many of the details of our images. However, the simplest and most intuitive model that reproduces the data requires only two electron wavefunctions and their elementary superpositions. Indeed, the success of our analysis is perhaps surprising in light of the well-known softness of the corral walls[19]. Without negating more complex approaches, this work vindicates the simple eigenmode picture of quantum corrals, which is relevant and useful to a variety of emerging applications.

For each superposition in Fig. 2, we selected the coefficients a_j to create $\Psi(\mathbf{r};\mathbf{R}) = \sum_j a_j(\mathbf{R})\left|\psi_j(\mathbf{r})\right\rangle$, where **R** is the atom position and **r** a position within the ellipse, subject to the normalization constraint $\sum_j \left|a_j\right|^2 = 1$. If only two wavefunctions participate for a given **R**, the state can be represented by a vector on a Bloch sphere: $\left|\Psi\right\rangle = \cos(\theta)\left|\psi_1\right\rangle + e^{i\phi}\sin(\theta)\left|\psi_2\right\rangle$. However, time-reversal symmetry requires that $\left|\Psi\right\rangle$ be real, so we are restricted to $\phi \in \{0,\pi\}$. Thus, we describe the mirage-projecting