

Probing Periodic Properties of “Artificial Elements” Assembled in a Quantum Wedge

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As an extension to the notion of an *artificial atom*, *artificial elements* are appropriate descriptions of artificial atoms with incremental number of occupied quantum states, for a succession of which can exhibit periodic properties in resemblance to Mendeleev’s Periodic Table of natural elements. As an example we show that an assembly of successive artificial elements can be realized through the fabrication of a *quantum wedge*. The energy level of the highest occupied quantum state in these elements display a two fold periodic shift which gives rise to a binary electron interference fringes, as imaged with a low temperature scanning tunneling microscope.

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An *artificial atom* [1] is a pool of electrons confined in a sufficiently small metal or semiconductor either by an energy potential barrier existing at the boundaries, or by a electrostatic potential introduced through a set of properly arranged electrodes. This is so named because both the charge and energy of such a small electronic system are quantized just like those of a natural atom. Artificial atoms, however, have their own unique properties, which often reveal atom-like features.

A natural atom is a chemical element with a characteristic electron shell configuration. The periodic repetition of electron configuration of a succession of elements give rises to the periodic repetition of their chemical properties as revealed most elegantly in Mendeleev’s periodic table. It is tempting, therefore, to ask whether an artificial atom could have its own identity that can be characterized by an *artificial atomic number* just like in the case of a natural atom?

In this paper we shall argue that *artificial elements* are a useful extension to the notion of artificial atoms for describing the latter with a small but distinct number of filled quantum states (QS). As a concrete example, we show that a succession of artificial atoms can be assembled in a metallic quantum wedge [2], which, indeed, exhibits periodic properties.

The number of filled QS and their energy spectrum of an artificial atom depend on its physical dimension (or size of the confining potential well). Consider for simplicity a one-dimensional metal artificial atom of size $H = Nd_0$, where N is the number of atomic sites and d_0 the interatomic spacing. The total number of QS in the partially filled conduction band is equal to N , and increases by one with the addition of each new atomic site in the artificial atom. Within a good approximation, the energy separation between two successive

QS near Fermi level, E_F , is [3]

$$\Delta = \pi \hbar v_F / H, \quad (1)$$

where v_F is the Fermi velocity and \hbar Planck's constant. For the highest occupied quantum state (HOQS) and the lowest unoccupied quantum state (LUQS), their energy shift when the size of the artificial atom changed from N to $N + 1$ is

$$\delta = \frac{2d_0}{\lambda_F} \Delta, \quad (2)$$

where λ_F is the Fermi wavelength of the conduction electrons. Eq. (2) asserts that the position of HOQS or LUQS relative to E_F shall exhibit a pattern of quasiperiodic repetition with a successive increase of N , in resemblance to the periodic properties of natural elements with N playing the role of atomic number Z . The period, however, is material dependent as reflected by the ratio of $\lambda_F/2d_0$. An example for a period of two is shown schematically in Fig. 1.

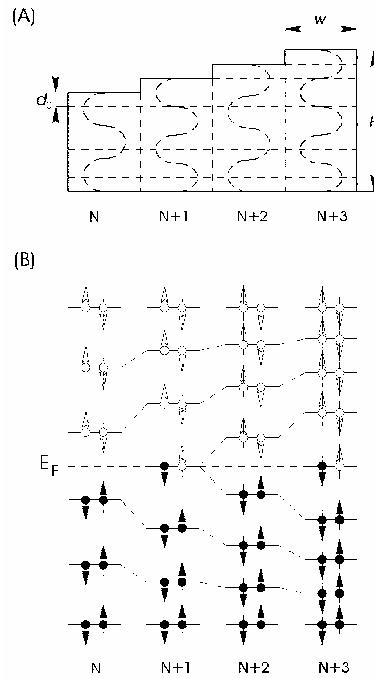


FIG. 1: (a) A quantum wedge consisting of four quantum wells with incremental atomic layers (the width is compressed disproportionately). The standing wave of the highest occupied quantum state for each element is illustrated. (b) Schematic of the energy level configuration for four successive number of total QS in an ideal half-filled band of a metal. It shows a quasi two fold periodic repetition near the Fermi level, i.e. $\lambda_F/2d_0 = 2$.

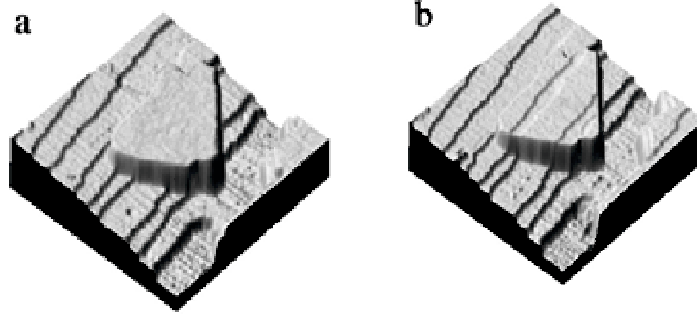


FIG. 2: (a) $5000\text{\AA} \times 5000\text{\AA}$ STM topography of a Pb wedge grown on a stepped Si(111) surface. The thickness of the wedge increases successively from 8 to 12 layers. (b) STM image of the same wedge taken with an opposite polarity of the tip bias (+5V), revealing the interference fringes of the highest occupied quantum state near the Fermi level.

With the advanced nano fabrication technology, it is possible to tailor the size of an artificial atom so that the number of QS can be adjusted by unity, hence create a periodic table of artificial elements. In particular, an array of artificial atoms with an incremental number of QS can be realized through the fabrication of a *quantum wedge*, i.e. a nanoscale metallic wedge whose thickness changes monotonically by discrete atomic planes. Each increase in the thickness by an atomic layer adds a new QS into the energy band as a result of matching the boundary conditions. Thus a quantum wedge is an assembly of artificial atoms with incremental sizes, or a succession of artificial elements.

To create the nano wedge described above, Pb islands of nanometer scale is grown epitaxially on a Si(111) surface with monatomic steps. A topographical image of a typical Pb wedge taken *in situ* by a scanning tunneling microscope at 5 K is shown in Fig. 2a. The island expands several atomic terraces of the substrate but its top is atomically flat except for a small vertical lattice mismatch. Thus as the Si substrate descends down from left to right, the number of Pb layer on each terrace increases successively from 8 to 12. This unique geometry results from the balancing act between the minimization of strain energy vs. the surface energy in this heteroepitaxy system.

Because of the large aspect ratio, the energy quantization in each element in the Pb wedge takes place only in the direction normal to the surface. Namely, the normal component of the wave vector $k_{\perp} = n\pi/H$, where n , being an integer, is the index of the n th QS. The energy spectrum for a given element is a set of discrete subbands for the electrons are free to move in planes.

Using site specific tunneling spectroscopy, a portion of the quantized spectrum for each of all the elements can be measured. Fig. 3 is a set of such spectra where the integer next to each of the curves indicates the number of Pb layers of that particular element. Note that due to the focusing effect of the tunnel junction, only states with a small in-plane wave vector k_{\parallel} in a subband contribute to the tunneling spectrum. Together with the weak dispersion near the [111] symmetry point of Pb [4], it gives rise to the sharp steps in the

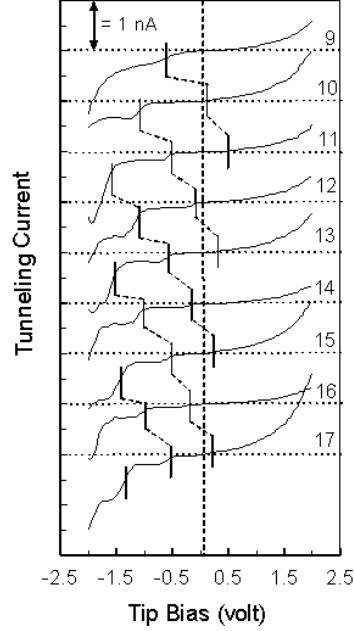


FIG. 3: A series of I-V spectra acquired at 5 K over an individual quantum element assembled in a Pb wedge. The integer next to each spectrum indicates the number of Pb layers in that element. Short vertical bars trace the progressing of quantum states with the same index.

tunnel spectra. In the region of negative tip biases, each step corresponds to a unoccupied QS, whereas in the positive region the dominant source of the tunnel current is from the HOQS. This asymmetric behavior is also an intrinsic property of the tunnel junction [2].

Fig. 3 clearly shows the progression of the n th QS with successive N as marked by the traces. Fitting the data of Fig. 3 to Eq. (1), we obtain a Fermi velocity $v_F=1.9\times 10^8$ cm/sec, in agreement with the bulk value of Pb [5]. It is also evident that the level of QS near E_F exhibits a two fold repetition. All the QS near E_F for all the *even elements* are more or less aligned, and so are those of *odd elements* (except for number 9 and 10). But between these two groups there is a substantial energy shift, and for the HOQS or LUQS of two successive elements, the shift is approximately $\Delta/2$. This is precisely what Eq. (2) predicts since for Pb, $\lambda_F=3.7d_0$ ($d_0=2.86$ Å), so that $\delta = 2\Delta/3.7$. It is even more remarkable that the pattern of the quasi two fold repetition is interrupted at $N=10$, just as expected from the irrational relationship between λ_F and $2d_0$.

The large shift of the energy spectrum between two successive elements makes it possible to probe the QS spectrum of an individual element with great clarity even though they are physically connected. The periodic nature of the energy shift gives rise to a periodic oscillation of the tunneling current at positive tip biases as evident in Fig. 3. When electrons tunnel from the wedge to the tip, those at the HOQS make dominant contributions and the closer the HOQS to E_F , the smaller the tunnel barrier. Thus all the odd elements yield

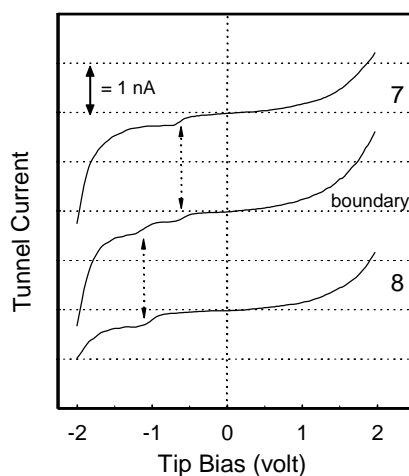


FIG. 4: I-V spectra acquired on two neighbouring fringes with a thickness of 7 and 8 Pb layers, respectively. The middle curve is a spectrum measured right on the boundary of the two fringes.

much higher current intensity than the even elements for $N > 10$, and the opposite is true for $N \leq 10$. Imaging the wedge under such condition, therefore, yield a marvelous binary fringe pattern as shown in Fig. 2b.

From Fig. 3 we can draw a somewhat unusual conclusion that even on the same metallic island the electron spectrum is different from one place to another, depending on the size of the structure down to atomic scale. This effect, generally known as the quantum size effect (QSE), is related not just to the size but also to the geometry as we have seen here. A more interesting question is what happens at the interface between two such artificial elements with their “ Z ”-number differed by one? Fig. 4 compares the I-V spectrum acquired right at the boundary (middle curve) with those taken on two adjacent terraces with a one layer difference in thickness. It shows that electrons from both sides retain their own energy all the way to the interface with no smearing. This is due to the large energy shift of the electron spectrum with the change of one atomic layer in the thickness. It implies that unlike the spatial confinement in the transverse direction, electrons are localized laterally within the strip of a particular Z -number due to transverse energy quantization rather than a physical boundary. This special effect could be utilized for building a QSE transistor [6].

As illustrated in Fig. 1B, element N and $N + 2$ have a “close shell” configuration and $N + 1$ and $N + 3$ have a “half-filled shell” configuration and are expect to exhibit insulating and metallic properties, respectively, in resemblance with the elements in the Mendeleev’s periodic table. Those with close shell configuration have a lower electronic energy than those with half-filled shell. Fig. 1 depicts an ideal case but as we have seen that Pb island on Si(111) turns out to be very close example of it. Indeed, it has been shown that when counting the Pb islands grown on Si(111) substrate, islands with odd number of layers (5, 7, 9) dominate the even number layer islands [7]. This is because the lower overall energy

and better stability of the odd layer islands (when $N < 10$) than the even layer islands. Furthermore, we have observed [8,9] that Pb atoms selectively grow on odd layer strip (for $N > 10$), bypassing the even layer strips, converting a island with equal number of strips for the odd and even layers to one only contains even number of layers. When the original higher energy configuration is converted to a lower energy one, the growth changes to a *double layer growth* on top the even layer strip so to preserve its lower energy configuration.

Like the natural elements in the Mendeleev's periodic table, the quasi-periodic repetition of QS configuration in a succession of artificial atoms can lead to repetition of its physical properties. One remarkable example is the manifestation that the normal to superconducting transition temperature of ultra thin Pb layers indeed oscillates between even and odd layers [10].

In an artificial periodic table, the period is determined by $\lambda_F/2d_0$. While d_0 is purely a material parameter, λ_F is associated with the percentage of the total QS that are filled, and therefore can depend on the size of the artificial atom as well as the valence electron density of the material. Since the energy dispersion is, in general, nonlinear away from the center of a band, a wide range of period for the repetition of the QS level can be expected for other materials and structures. Indium has one less valence electron than lead, and we found that the period for In(111) thin layers grown on Si(111) equals 4. It should be noted that Eqs. (1) and (2) are no longer applicable in the nonlinear regime. Finally, a first principal calculation has confirmed the repetitive character of the QS of thin Pb layers [12].

In summary, using a Pb quantum wedge as an assembly of artificial atoms with a successive "Z-number", we have demonstrated the existence of a quasi-periodic repetition of the physical properties of these artificial associated with the repetition of the QS configuration, and have thus shown the worthiness of the notion of "artificial elements" as an extension to that of an artificial atom.

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