

Approaching the Ballistic Transport Limit in Single Crystalline Au

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Quantum conductance effects start to appear as the size of electronic devices decreases and they are used in the next generation memristors and quantum calculations. Understanding the relationship between line width and resistance is crucial for developing quantized electronic devices. This Letter reports the development of an electromechanical testing device based on microelectromechanical systems integrated within a transmission electron microscope for *in situ* investigations at atomic resolution. The resistance changes of single crystalline gold (Au) wires, spanning from hundreds of nanometers down to several atoms are investigated. A transition in resistance from quasiballistic to ballistic transport states is uncovered. The experimental data fitted by calculation show that the ballistic transport limit for single crystal Au at room temperature is about 2.5 nm, beyond which the transmission probability experienced a rapid drop.

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Introduction—Numerous concepts for devices utilizing quantized conductance have been proposed. When the size is reduced to several atoms, the quantum conductance effect can manifest even at room temperature. This phenomenon was first discovered in semiconductor devices in 1990 [1]. And the quantized conductance atomic switch (QCAS), proposed in 2005, has attracted wide attention due to its highly controllable quantum conductivity, high switching speed, and low energy consumption [2]. These attributes position devices exhibiting quantized conductance as promising candidates for the next generation of non-volatile memory and logic operation circuits [3,4].

The concepts utilizing quantized conductance were proposed [5,6], revealing substantial potential applications in areas such as synapse emulation and photon detection [7,8]. However, the operation of these devices fundamentally rely on the quantum conductance effect, which arises from the ballistic transport mechanism at the atomic scale. A critical challenge in this domain is that higher conductance is not as easily controllable as its lower counterparts [2]. This phenomenon is attributed to the increasing

probability of electron scattering as conductor sizes increase, resulting in a transition from ballistic transport, characterized by a transmission probability $T = 1$, to quasi-ballistic transport, with a transmission probability $0 < T < 1$. Such a transition can lead to fluctuations in conductance and the emergence of noninteger values [9]. Therefore, it is important to explore the size limitations of quantized conductance devices.

The conductive behavior of metallic materials can be categorized into three stages based on the comparison between the sample size with the electron mean free path. When the sample size d is significantly larger than its electron mean free path l_e , electron transport occurs in a diffusion mode. And when $d \ll l_e$, the transport mechanism transitions to ballistic transport [10]. The intermediate region, characterized by $0 < T < 1$, is referred to as the quasiballistic transport region, where the transmission probability T plays a crucial role. During the ballistic transport, the conductance of the conductor follows the Landau formula $G = G_0 \sum_{n=1}^N T_n$, in which $G_0 = 2e^2/h$, and e represents electron charge; h stands for Planck's constant; T_n refers to the transmission probability of the n th channel [11,12]. In these situations, variations in conductance are manifested as discrete increments of fixed units G_0 [12]. This characteristic is designed for next-generation logic arithmetic and memristor applications. However, the dimensional limits of ballistic transport remain undetermined, and the behavior of the transmission probability T

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changes within the quasiballistic transport region is still not fully known.

To investigate the size effects of devices exhibiting quantized conductance, pioneer research has explored electrical transport mechanisms [13–15]. Techniques such as scanning tunneling microscopy (STM) and mechanical controllable break (MCB) methods enable the fabrication of ballistic nanowires with dimensions on the order of a few nanometers, facilitating measurements of electrical signals related to ballistic transport [16–25]. However, concerns may arise regarding heating effects and the limitations of two-electrode measurements. A desired consideration in measurement technology is the avoidance of heating and using the Kelvin method [26] which significantly improves the resistance measurement accuracy compared to the two-electrode method, by mitigating contact resistance artifacts and improving contact stability.

Recent developments in *in situ* transition electron microscope (TEM) techniques have enabled real-time structural observation [13–15,27,28], facilitating investigations into the influence of local atomic configurations on electrical parameters [13–15]. However, the probe-based experimental methods have been meeting challenges using Kelvin method measurements in addition to the difficulties of handling large-sized samples. The underlying mechanisms governing conductance variations ranging from hundreds of nanometers to the atomic scale remain incompletely understood. Within this range, the transport mechanisms include diffusive transport, quasiballistic transport, and ballistic transport. It is essential to study conductance variations across different width scales and delineate the critical dimensions that define the limits of ballistic transport [3]. As the electrical transport mechanism transforms, resistance may exhibit changes from a linear to a nonlinear response, or it may follow a fixed value growth pattern. It is important to determine the variation trend of electrical properties with size and the underlying transformations in the electrical transport mechanisms. This knowledge will help to determine the limiting size for ballistic transport and guide the design of a new generation of quantized conductance devices.

In this Letter, we present a novel electromechanical coupling microelectromechanical systems (MEMS) based double tilt sample holder for TEM. The innovative design allows the simultaneous measurement of local electrical signals using the Kelvin four-electrode method and real-time atomic-resolution observation, while avoiding sample heating. This setup accommodates samples with sizes ranging from several micrometers down to several atomic dimensions. By conducting a uniaxial tensile test on single crystal Au beams, we successfully achieved real-time measurements of electrical signals and image observations ranging from hundreds of nanometers down to the atomic level. The results led to the generation of a conductance-width curve, which highlighted a potential transition point

from quasiballistic transport to ballistic transport. Notably, our findings indicate that the critical size of ballistic transport at room temperature is about 2.5 nm. Exceeding this size the transmission probability experienced a rapid drop.

Method—MCB is a widely used method for achieving atomic contacts with exceptional controllability and precision in electrical measurements. Nevertheless, acquiring high-resolution images remains a challenge. To address this limitation, a MEMS-based MCB technique that can be applied in TEM was developed. A mini laboratory setup was created, incorporating a piezoelectric actuator, a MEMS device, a support structure and a double tilt mechanism, as shown in Fig. S1(a) [29]. This mini lab applies in-plane load through a piezoelectric element, while the electrical signals of the sample were transmitted and measured through four wires integrated into the MEMS chip, as illustrated in Figs. S1(b) and S1(c) in the Supplemental Material [29].

Focused ion beam (FIB) technology was utilized to fabricate miniature tensile samples of single crystal Au with a [110] normal orientation within the mini lab. The samples were thinned down to approximately 100 nm and then graphically shaped to achieve beam samples with a dimension of 400×1200 nm, as depicted in Fig. S1(d). The device operates in conjunction with a specialized double tilt sample holder, which allows high resolution imaging during the tensile process, as illustrated in Fig. S1(e) [29].

The system was utilized to conduct tensile experiments on single crystal Au, with widths ranging from 400 nm to a few atomic widths along the approaching direction $[1\bar{1}2]$. The deformation control achieved by the micropiezoelectric ceramic driving device was less than 0.1 nm [34]. The driving force can be applied through a group of piezoelectric ceramics at the front end of the MEMS, which is the PL022.30 PICMA[®] produced by PI. When the voltage is applied to the piezoelectric ceramic, it expands along the tensile axis due to the piezoelectric effect, shown as Fig. S2(a), which drives the mass block supported by the multibeam structure on the MEMS device to move. The compression displacement of the input is transmitted to the push-to-pull sample area through the constraints of multiple beams in a single axis to realize the stretching of the sample, shown in Fig. S2(b) [29]. By controlling the voltage applied to the piezoceramic driver, high-precision displacement can be achieved, with a drive bit shift of about 0.2 nm when the input voltage step is 0.1 mV, shown in Fig. S2(c) [29]. Because of the good plasticity, Au would be necked during the stretching process, resulting in continuous changes in width, as depicted in Figs. S1(f) and S1(g) [29]. Moreover, the four-terminal sensing measurement method simultaneously eliminated the influence of external circuits and MEMS device electrodes, resulting in a more accurate current signal [28]. This experiment setup achieves a measurement accuracy of 10 nA under input voltages at μ V level, thereby

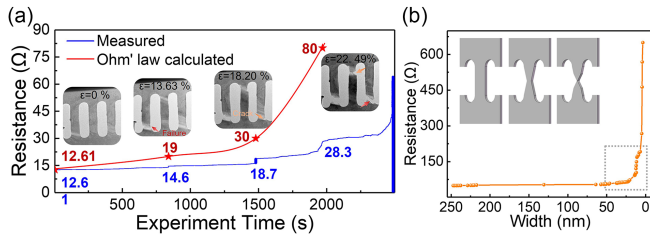


FIG. 1. Variation between width and resistance. (a) Experimental resistance data and resistance calculated by Ohm's law. (b) Measured width was related to the resistance.

significantly minimizing the effects of electromigration and temperature elevation that are typically associated with high-voltage measurements [28,34,35], as shown in Supplemental Material Fig. S3 [29]. Throughout these experiments, both images and current signals were recorded simultaneously.

Experiment and result—The experiments revealed that the electrical signal deviated from Ohm's law in multiple beam samples, with the degree of deviation increasing as the total cross-sectional area decreased, as shown in Fig. 1(a). Three beams were adopted in the experiment, with an initial total resistance measured at approximately 12.61 Ω. A current-time curve was recorded during the complete stretching-to-fracture process. At a tensile strain of 13.63%, the left beam fractured, which resulted in a rise in resistance. When the tensile strain reached 18.2%, cracks emerged at the root of the right beam, which led to a noticeable increase in resistance. When the tensile strain reached 30%, the right beam completely broke, causing a 58% reduction in the width of the middle beam. Compared to the initial sample, the total cross-sectional area of the sample was reduced by at least 86%, yet the resistance increased by only 1.24 times, which significantly lowered than the sevenfold increase predicated by Ohm's law.

To clarify this variation, the experimental resistance data were graphed as blue line. The theoretical resistances at three pivotal points were calculated using Ohm's law, considering the parallel relationship between beams and the resistance drop due to width reduction, represented by a red star in Fig. 1(a). The accompanying TEM images demonstrate that the correlation between changes in resistance and Ohm's law deteriorated as the beams fractured and the cross-sectional width diminished. This deterioration in correlation can be attributed to the size reaching the transition region between diffusion and ballistic transport. As the size of the beams decreased, the number of electrons transported by ballistic means increased, while scattering events became less frequent. Consequently, the resistance rose at a slower rate than what would occur under diffusion transport conditions.

To expound the impact of section width on resistance variation, the relationship between width and resistance was examined for a single beam sample, as depicted

in Fig. 1(b). It was observed that minimal change took place in resistance when the width decreased from the 100 to 20 nm connection during the sample shrinkage and rupture process, with only an increase of 20% observed. Given that the mean free path of Au at room temperature was approximately 37.7 nm [36], the minimum dimension of the sample in the three-dimensional space was its thickness, which measured about 100 nm. This implies that the width of the electron potential well was comparable to the average free path of electrons. In this case, the transport mode sat between diffusion and ballistic transport, with the transmission probability of electrons being less than 1 [9]. As shown in Fig. 1(b), a significant increase in resistance was observed at widths below 20 nm. Through the derivation of the curve, it can be found that the jump point of derivative value growth was about 20 nm. When the sample width was greater than 20 nm, the resistance growth rate remained lower than what would be expected under only diffusion transport conditions. When the width of the necking region was lower than the mean free path, the electron transmissivity in quasiballistic transport began to increase slowly, approaching 1. At this juncture, the resistance at this point was mainly controlled by the number of conductive channels available. As the cross section decreased, the resistance increased in the form of a square.

High resolution transmission electron microscopy (HRTEM) was utilized to investigate the alterations in electrical signals before the sample fractured at multiple atomic junctions. The objective was to identify the critical dimensions at which the sample transitions from quasiballistic transport characterized by scattering to a ballistic transport mode. An atomic junction with a width of several nanometers was stretched at a rate of 1 nm/s, with corresponding images and resistance values recorded in real time, as shown in Supplemental Material Fig. S4 [29]. Following the tensile fracture, the atomic junction could be repeatedly formed through processes such as cold welding [37] or pulse welding [38], as shown in Fig. S5 [29]. This enabled the calculation of the relationship between the width of the atomic junction and its resistance. The width-resistance values were recorded and fitted using an exponential function of type $R = A^{(W+B)} + n$, as depicted in Fig. 2(a). The fitting result was $R = 0.24^{(W-6.37)} + 257$. To understand the sensitivity of resistance to changes in width, the derivative of the fitting function was calculated, revealing the change rate of resistance with respect to width. Notably, the growth rate of resistance was found to be 1 Ω/nm at a width of $W = 6.6$ nm, while it reached 100 Ω/nm at $W = 3.4$ nm, indicating that the device can achieve a large switching ratio.

To more accurately determine the transition point of the conduction mechanism, the resistance values obtained from the experiment were converted into conductance and divided by the unit value G_0 of the conductance quantum

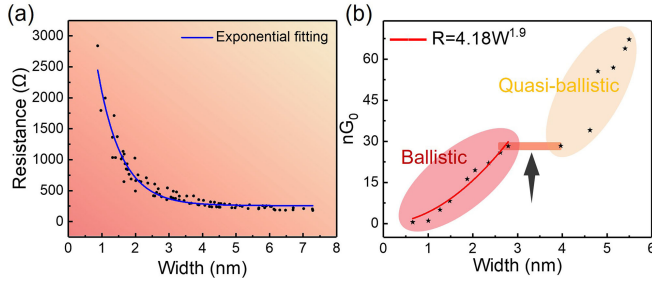


FIG. 2. Relationship between the width and electrical signal of the nanoscale sample. (a) Statistical resistance-width curves and fitting results. (b) Width-quantized conductance relation.

to determine the conductance quantum number n of the sample, as illustrated in Fig. 2(b). It was observed that the conductance of data points below 2.5 nm exhibited a well-defined square relationship with width, whereas data points above 2.5 nm did not display a similarly pronounced trend. In the context of ballistic transmission, the conductance was determined by the number of transmission channels in Au, which corresponds to the number of atomic chains present [16,27,39]. At the nanoscale, surface free energy effects would cause the minimization of contact section, resulting in a reduction to circular or square geometries to minimize energy [40]. For rectangular or cylindrical cross sections, the relationship between the diameter squared and conductance is more consistent, particularly for widths below 2.5 nm. Similarly, a small platform between 2.5–4 nm indicated that the transmission mode may have shifted from quasiballistic transport, characterized by scattering, to a ballistic transport mode that occurs without scattering.

When a slower drawing rate of about 0.1 nm/s was used, a clearer conductance step was obtained. Taking into account potential measurement errors arising from the electron beam interactions [41–43], we systematically evaluated the mechanical, heating and additional electrical effects induced by the electron beam might have under the experimental conditions, as shown in Fig. S6–S8. The results indicate that the influence of the electron beam is negligible. To facilitate a more accurate observation of conductance quantization, the ordinate was expressed in terms of the quantized conductance unit G_0 , as depicted in Fig. 3(a). The figure illustrates the relationship between the number of atoms, width, and conductance. Additionally, a series of simultaneously collected images are comprised in Fig. 3(b). The conductance exhibited significant quantization behavior upon reaching 43 G_0 , with the corresponding atomic width being fewer than 14 atoms. Notably, the quantized values were not integers. According to the findings reported by Torres and Sáenz [44], integers were typically quantized exclusively in the systems containing only a final few atomic chains. This indicates that the elasticity fluctuations at the contact point and the

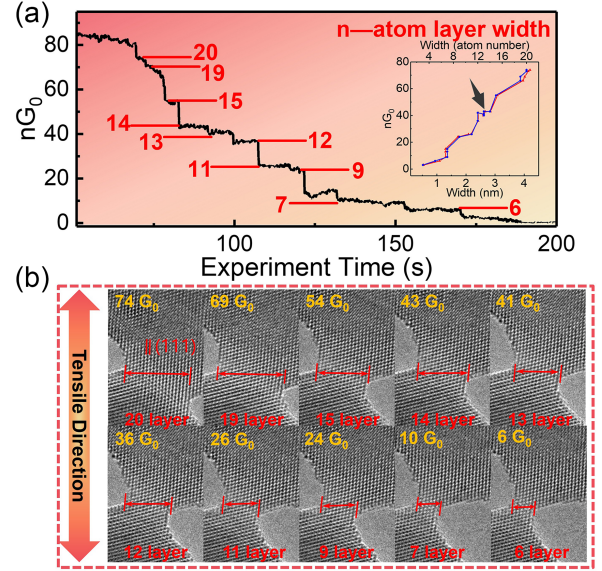


FIG. 3. Quantization of conductance at low width dimensions. (a) Conductance-width curve. (b) Corresponding image of the conductance-width curve.

rearrangement of the local atomic structure can lead to fluctuations within a small range [45].

In the illustration, a region of approximately 2.5 nm also corresponded to a plateau where the absence of two columns of atoms exerted a minimal impact on conductance. During the tensile process of atomic contact, two factors affected the changes in conductance. The first factor was the decrease in size, which reduced the cross-sectional area or the number of conductive channels, leading to a decline in the overall conductance value. The second factor involved the electrical transport mode, which approached ballistic transport; as the size decreased, the scattering probability of electrons decreased, resulting in an increase in overall conductance. The presence of plateaus indicates a balance between these two influence factors. The observed phenomenon suggests a transition in the transport process within the sample, shifting from quasi-ballistic to ballistic transport in a scattering-free state. As the size continued to decrease, the narrowing of the transmission channel occurred, yet the transmission probability stayed at 1, leading to a further decrease in conductance. As the size became smaller, the atomic structure became increasingly unstable and tended to break rapidly during the stretching process, without capturing the corresponding image of the atomic arrangement.

To investigate the transition point of the conductive mechanism, the modified Sharvin formula (ballistic transport) [46,47] was used to compute the resistance at the contact point in the experiment. For the purpose of these calculations, a cylindrical shape was used and confirmed (see Fig. S9 and corresponding text in Supplemental Material [29]) for the contact point. This implies that

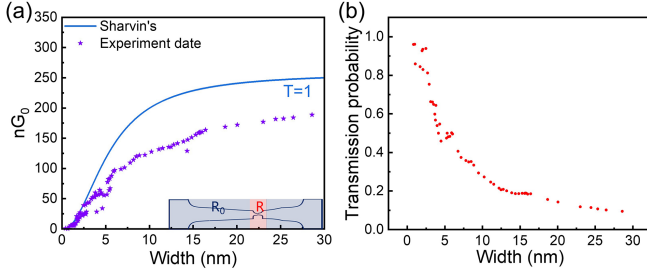


FIG. 4. Calculated conductance and transmission probability. (a) Comparison of calculation and experimental data using the modified Sharvin formula. (b) The relation between transmission probability and width obtained by experiment.

the alignment of atomic columns in the Z direction was consistent with the width direction.

$$G_c = TG_0 \left(\pi \frac{S}{\lambda_F^2} - \alpha \frac{P}{\lambda_F} \right). \quad (1)$$

Where G_c is conductance, T is transmission probability, for ballistic transport $T = 1$. G_0 stands for quantum conductance unit, λ_F is Fermi wavelength of gold, S is the section area, P is the section perimeter, and α refers to the shape factor, with $\alpha = 0.5$. Given that potential ballistic transport was limited to the atomic junction on the order of a few nanometers, stretching the rest of the sample introduced an initial resistance, as illustrated in Fig. 4(a). According to the experimental results presented in Fig. 1, the initial resistance of $R_i = 50 \, \Omega$ was added in parallel with the atomic junction part to calculate the total conductance G_{cal} . The conductance quantum number was calculated by $n = G_{cal}/G_0$:

$$n = \frac{9.15d^2 - 3.02d}{1 + 0.003875(9.15d^2 - 3.02d)}. \quad (2)$$

The detailed derivation process can be found in the Supplemental Material [29]. The calculation results based on the modified Sharvin formula, which represents ballistic transport is plotted in Fig. 4(a). In this figure, the purple stars represent experimental data points. It is observed that the experimental data aligns with the ballistic transport results for sizes below 2.5 nm. However, as the size exceeds this threshold, the data begin to deviate from the expected ballistic behavior, which indicates that once the size surpasses the critical threshold, scattering will intensify, leading to a decrease in the proportion of ballistic transport. This observation provides evidence that the transformation from ballistic to quasi-ballistic transport occurs at the size of 2.5 nm.

By subtracting the initial resistance from the experimental data, the conductance of the atomic junction can be obtained. This conductance is then normalized by dividing it by the conductance of the atomic junction at $T = 1$,

allowing for the investigation of the transmission probability of the atomic junction, as shown in Fig. 4(b). The results reveal that T approaches 1 for junction sizes below 2.5 nm, while T decreases rapidly when the size exceeds 2.5 nm. This decline suggests that $T \neq 1$ less than 2.5 nm is due to the stress and structural fluctuation on the junction, which causes fluctuations within a small range.

Summary and outlook—This Letter presents a groundbreaking real-time detection of resistance changes and imaging of samples ranging from hundreds of nanometers down to atomic-scale widths. The variation trend in resistance with width deviated from Ohm's law, revealing that the resistance growth rate is relatively slow above 20 nm, but grows rapidly below 20 nm, reaching a remarkable growth rate of 100 at 3.4 nm. The data on the rate of resistance change offers valuable experimental insights for understanding the switching ratios of quantized conductance devices. Through atomic-scale observations and measurements, we demonstrate that the transport state gradually changes from quasiballistic to ballistic transport state. A comparison of calculated and experimental data indicates that samples below 2.5 nm conform the calculated results of ballistic transport. In contrast, the electron scattering becomes evident when the sample width exceeds 2.5 nm. The experimental results of transmission probability T further support this finding, showing a sharp decline above 2.5 nm, which suggests a size limit for ballistic transport in Au at room temperature. This research will provide reference for future design considerations in the functional unit sizes of quantized conductance devices, including memristors and logic arithmetic devices.

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X. Y., J. Z., and W. L. contributed equally to this work, they conducted the experiments and wrote the initial manuscript. Q. Zhang and M. W. assisted in the design and construction of the experimental setup. Q. Zhou and S. Z. helped with sample preparation and data analysis. S. M., L. W., and Z. Z. designed and guided the research details. X. H. conceived the idea and supervised the project. All the authors reviewed and revised the manuscript.

Data availability—The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be

prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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Correction: Author contribution statements have been added.