Quantization effects in the conductance of metallic contacts at room temperature

C.J. Muller
Department of Electrical Engineering, Yale University, P.O. Box 208284, New Haven, Connecticut 06520-8284

J.M. Krans
Kamerlingh Onnes Laboratory, University of Leiden, P.O. Box 9506, 2300 RA Leiden, The Netherlands

T.N. Todorov
Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, England, United Kingdom

M.A. Reed
Department of Electrical Engineering, Yale University, P.O. Box 208284, New Haven, Connecticut 06520-8284
(Received 30 May 1995)

We present measurements on conductance quantization in atomic scale metallic constrictions at room temperature. Conductance measurements as a function of constriction size reveal pronounced horizontal plateaus separated by sharp discrete steps. Most individual conductance plateaus are close to integer multiples of the fundamental conductance unit but definite deviations are also present. After averaging over many contact configurations, conductance quantization is seen clearly and more prominently than in similar recent low temperature measurements on the same metals.

Few effects reveal fundamental constants in transport measurements, and experiments which bring out these constants are of fundamental importance. In recent years, considerable attention has been focused on conductance quantization in controllable two-dimensional electron gas (2DEG) point contacts after the initial discovery by van Wees *et al.* and Wharam *et al.*[1] As a natural next step a number of people have tried to produce small controllable metallic contacts in order to investigate whether these systems (with a much higher Fermi energy as compared to the 2DEG systems) also exhibit the quantization effect.[2–5] The results in the latter case, however, are more complex and their interpretation is less straight-forward. The reason is that while a number of experiments show that the transition from contact to tunneling—where the electrodes lose contact—takes place at $2e^2/h$, measurements in the contact regime do not always show clear horizontal conductance plateaus at integer multiples of $2e^2/h$ when the constriction size is varied. Often conductance steps are observed which are not of the "right" size and which link noninteger conductance plateaus. Recent work has indicated that conductance quantization, if at all present, becomes apparent only after averaging over many contact configurations. This has been shown clearly in measurements on sodium at 4.2 K (Ref. 6) and also in room temperature work (see Ref. 7).

A related experimental problem is the stability required to maintain an atomic scale contact constant in size. Often conductance traces have to be recorded in milliseconds or less since the contact changes its size on this time scale. Some experiments show gradual transitions between conductance levels, possibly resulting from this small recording time, while other experiments show abrupt discrete steps. This has been the basis for some controversy in the past since an abrupt conductance step favors a different physical mechanism from a gradual transition.[7]

"Conductance quantization" implies that the conductance is quantized in the fundamental unit of $2e^2/h$. This quantum effect is of similar importance as the quantized Hall resistance in the quantum Hall effect and the quantized voltage increments in the ac Josephson effect in the presence of radiation; the latter effects are accurate to a very high degree of precision and are therefore useful as standards. An experimental claim of conductance quantization must be accompanied by a range of validity and an explanation should be provided for the obtained level of precision. In this paper we show that the concept of conductance quantization in small metallic constrictions requires refinement. Our experimental results show that the quantization effect in such systems can be very prominent but is nonetheless not universally present. Comparison with past measurements shows that the conditions for the effect improve with increasing temperature. A firm understanding of the deviations from conductance quantization in these systems is of fundamental importance. We discuss specific mechanisms for these deviations. The experimental data are in qualitative agreement with recent theoretical results by Bratkovsky, Sutton, and Todorov.[8]

Contacts of gold and copper (99.99 %) are adjusted at room temperature with the use of the mechanically controllable break (MCB) technique.[9] A metal wire is glued on a flexible substrate; it is fractured by bending the substrate after which adjustable atomic size contacts can be established. This system allows for multiple contact formations and deformations. The main operating principle is shown in the inset of Fig. 3. It relies on a large reduction factor between the piezoelongation and the electrode separation. The small section between the two glue contacts ensures inherently stable contacts or tunnel junctions. In contrast to previous work

0163-1829/96/53(3)/1022(4)/$06.00

with MCB junctions these experiments are performed at room temperature. In order to reduce surface contamination the electrode material is fractured in a vacuum system (10^{-7} Torr), which uses an oil-free absorption/ion pump combination. Two-terminal conductance measurements are performed by biasing the junction at 26 mV and recording the current during a variation of the contact diameter. No qualitative difference in the recordings was observed by lowering the bias voltage. After the fracture of the metal wire, the thus created electrodes are carefully brought closer together in the tunnel regime, until suddenly a large contact with a typical resistance of 100 Ω is formed. This jump to a large contact allows us to perform only conductance measurements with decreasing constrictions. In our experiments the large contact cross section is reduced in size by increasing the piezovoltage until the conductance is about ten times $2e^2/h$. At this stage a long neck is drawn between the electrodes.[3] The piezovoltage is now fixed and the contact is allowed to relax by itself. The conductance decreases spontaneously in time, presumably due to surface diffusion of atoms away from the contact (i.e., the neck becomes thinner over time). This effect is shown for two contacts of gold and copper in Fig. 1 in the last stages of the contact while the conductance is measured with a sample rate of 100 Hz. Remarkably it appears that the conductance attains specific values near $n \times 2e^2/h$, with n an integer. Transitions between the levels occur abruptly. At some point the neck becomes so thin that the two electrodes can be bridged by a last single atom after which a jump to the tunnel regime occurs and the conductance becomes almost zero on the scale shown. This jump usually occurs near $n = 1$, and sometimes near $n = 2$ or 3. The length of the plateaus may differ for the various conductance traces. In general, however, we observe that copper contacts relax into the tunnel regime within 4 s, and gold contacts within 9 s. Some plateaus show structure and intrinsic noise which considerably exceeds our measurement accuracy. The noise amplitude is observed to increase for higher n.

The integer conductance values of the various horizontal plateaus such as those in Fig. 1 suggest the presence of conductance quantization in Au and Cu at room temperature. Caution is needed, however, in claiming this observation on the basis of individual traces, because apart from these integer values, in most conductance traces plateaus can be found which are off, sometimes even halfway, an integer value [see, e.g., panel (c) in Fig. 1]. The abrupt steps between the plateaus are caused by sudden atomic rearrangements in the constriction.[2,6,7,10] In each individual scan the precise atomic geometry of the constriction is different and the scans do not reproduce in detail. In order to give clear evidence for conductance quantization in these constrictions, we must show that the integer conductance values prevail over noninteger ones. To this end, a summation is performed over a large number of conductance traces, thus sampling a wide range of individual contact geometries. The only selection criterion for the traces was that the contact should relax into the tunnel regime within 4 s for copper and 9 s for gold. Such a criterion was necessary because it was noticed that typically 1 h after initial fracture the traces became longer or the conductance remained constant. At the same time arbitrary conductance values between $n = 0$ and 1 were observed and it was possible to adjust the conductance to almost any value. We attribute these effects to contamination of the electrode material by adsorbates which are forced into the material by repeated contact formations and deformations. The observed linear I-V curves of the selected contacts give us confidence that these contacts are metallic.

A total of 72 gold and 78 copper measurements (four samples of gold and three of copper) have been used to construct a suitable sample set. A histogram of the observed conductance values is shown in Fig. 2. The histograms of the individual samples are very similar to the ones shown here. Pronounced peaks around integer values of $2e^2/h$ are evident. Gold shows a narrow $n = 1$ first peak clearly separated from $n = 2$. For $n > 2$ a background starts to rise, but a number of peaks for $n > 2$ can still be resolved clearly. Copper behaves similarly

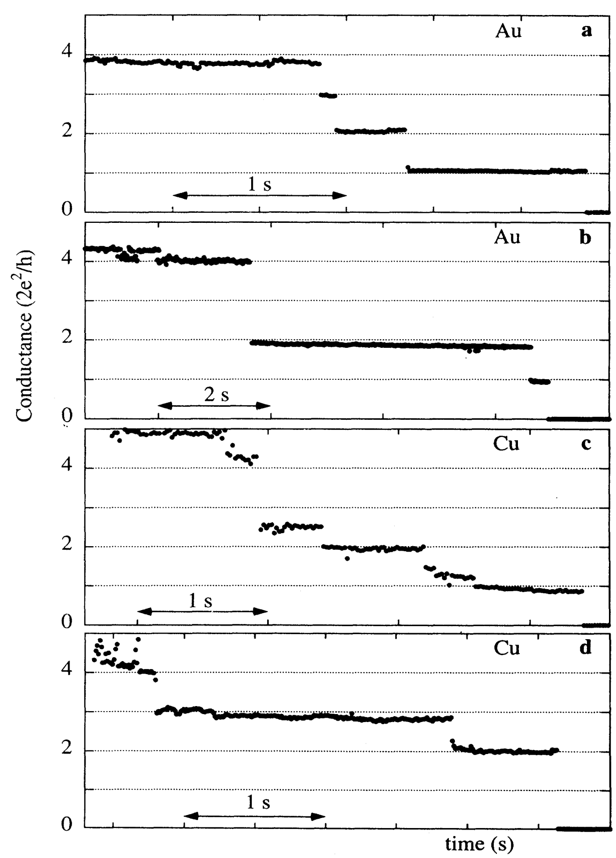

FIG. 1. Conductance traces of gold, panels (a) and (b), and copper, panels (c) and (d), showing clear plateaus separated by abrupt discrete steps. After adjustment of a contact with a conductance of about ten $2e^2/h$, the contact spontaneously reduced its size. During the displayed time interval the junction evolved from a few atom point contact to a tunnel junction after breaking. Both plateaus near integer values and plateaus far off are shown to be present.

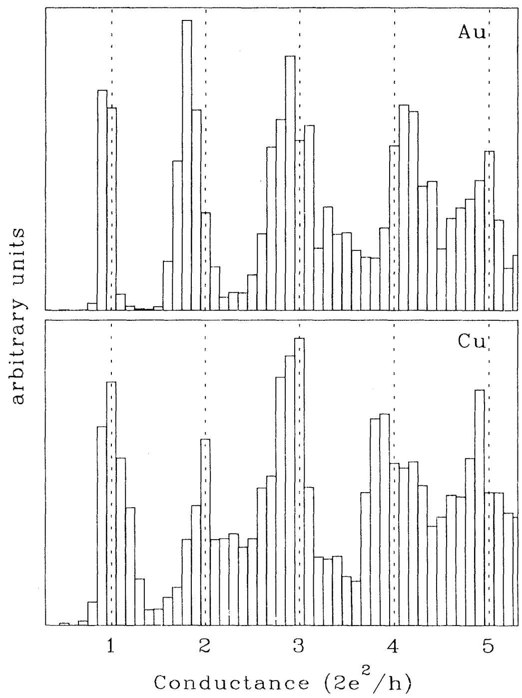

FIG. 2. The histograms of gold and copper reflect data of many conductance traces like the ones in Fig. 1. Both histograms are obtained from several MCB junctions. The fundamental unit $2e^2/h$ is divided into ten sections; the total number of data points of all traces in a specific section is represented by a bar. From this figure it is clear that there is a preference for the quantized values.

to gold, although the first peak is somewhat wider and not completely separated from $n = 2$. These histograms show that even in the absence of exact quantization in individual traces, the effect can be recovered by averaging over many different contact configurations. In contrast to the present room temperature results, in recent MCB experiments on Cu at 4.2 K, similar averaging revealed peaks only near $n = 1$ and 3 and additional smaller peaks at noninteger conductance values.[6,11] The width of the peaks shows that many plateaus occur at values just below and above $n \times 2e^2/h$, suggesting that the deviation from integers occurs randomly. However, a limited number of traces can be mapped exactly onto integers for multiple plateaus by the subtraction of a constant resistance value. This is shown in Fig. 3 where the plateaus $n = 2$ and 4 fall exactly on the integer values after 315 Ω is subtracted from all data points. A similar correction was used in the experiments on 2DEG semiconductor devices.[1]

So far, only the $n < 5$ conductance region has been discussed. The behavior for larger conductance values is shown in Fig. 4 for a trace of gold and copper. As in Fig. 1 an increase in noise is observed for larger n. At these larger values the noise amplitude can easily exceed $2e^2/h$, which prohibits the assignment of specific plateau values. Often, switching behavior occurs between two

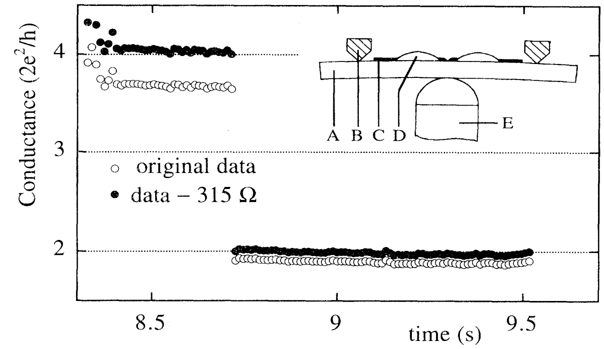

FIG. 3. For a number of traces multiple plateaus can be mapped onto integer values by subtraction of a constant resistance from the data. This finding is in accordance with similar experiments in 2DEG's. The inset shows a schematic drawing of the MCB junction with A the bending beam, B the counter supports, C the notched gold or copper filament, D the glue contacts, and E the piezoelement.

neighboring conductance levels. Another type of noise is observed to be more random (see left of the $n = 7$ plateaus in the copper trace). Noise due to the thermal vibrations of the atoms will have a time period of 10^{-13} s and will not be resolved experimentally. Plausible sources of noise are fluctuations in the geometry whereby one or a few atoms switch between energetically equifavorable configurations. This mechanism will depend strongly on temperature, consistent with the absence of noise at low temperatures.[2]

We define conductance quantization as a conductance staircase with plateaus at integer multiples of $2e^2/h$,[11] as opposed to simply conductance jumps between arbitrary values. In the case of a perfectly homogeneous

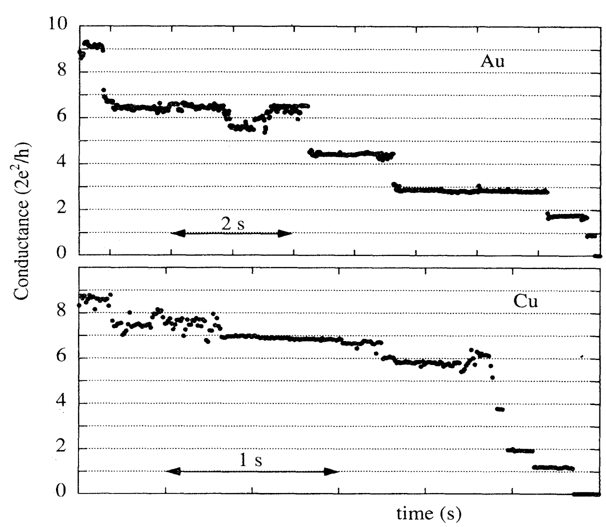

FIG. 4. These two conductance traces illustrate that the noise level at a plateau increases for larger n.

constriction of adiabatically varying cross section, each electron propagating along the constriction stays in a particular transverse state (channel). In this idealized system the transmission coefficients for all incident channels are either zero or one, and a quantized conductance of $n \times 2e^2/h$, where n is the number of propagating modes in the narrowest part of the constriction, then follows from the Landauer formula.[12] Small imperfections in the contact geometry will cause channel mixing, whereby electrons in the constriction undergo scattering between different propagating channels traveling in the same direction. In that case the total conductance will still be $n \times 2e^2/h$, even though the transmission coefficients for individual incident channels may be fractional.[13] In the presence of larger imperfections, backscattering (transitions between states traveling in opposite directions) may develop. Backscattering generally causes departures from conductance quantization. In the jellium picture, backscattering may result from nonadiabaticity and irregularities in the confining profile of the constriction. In reality, backscattering may also result from defects in the internal atomic structure of the contact. Theoretically, internal disorder has been studied only recently within a tight-binding model.[8,10]

None of our traces show conductance quantization in the strict sense, defined above. However, certain trace segments come very close to showing the effect, such as the last three plateaus in panels (a) and (b) in Fig. 1. At low temperatures, no comparable horizontal plateaus at integer conductance values larger than $n = 1$ have been observed in individual traces.[2,6] Furthermore, the present room temperature histograms show a number of peaks at integer conductance values, in contrast to corresponding low temperature results on the same metals.[6] At any temperature, in its mechanical evolution the contact seeks energetically favorable geometries. At high temperatures the higher kinetic energy of the atoms enables the contact to explore a wider range of structures in its search for the most favorable geometry. Recent theoretical work[8] shows that this has two important consequences. First, the contact tends to become more uniform in cross section and shape. Second, internal structural defects, appearing during the evolution of the contact, heal more rapidly, improving the crystallinity and internal order in the contact. Both effects improve the conditions for conductance quantization and explain the experimental observation that quantization occurs more readily at elevated temperatures.

In conclusion, our room temperature results show clearer signs of conductance quantization in individual traces than seen in previous measurements. After averaging over many contact configurations the effect at room temperature appears to be different from that at low temperature, especially for $n > 1$, where more peaks are observed at integer conductance values. But we also find persistent deviations from the effect in individual traces. These deviations carry important information about the structure of small metallic contacts.

We thank B.J. Vleeming for his assistance in building the high vacuum system. We acknowledge A.A. Verheijen and J.M. van Ruitenbeek for improvements to the manuscript. T.N.T. thanks his co-workers, A.M. Bratkovsky and A.P. Sutton, for authorizing him to represent their joint theoretical work at the Materials Modelling Laboratory at Oxford which is partially funded by the EPSRC under Grant No. GR/H/58278. C.J.M. gratefully acknowledges the Niels Stensen Foundation for financial support. We acknowledge ARPA for funding.

[1] B.J. van Wees, H. van Houten, C.W.J. Beenakker, J.G. Williamson, L.P. Kouwenhoven, D. van der Marel, and C.T. Foxon, Phys. Rev. Lett. **60**, 848 (1988); D.A. Wharam, T.J. Thornton, R. Newbury, M. Pepper, H. Ahmed, J.E.F. Frost, D.G. Hasko, D.C. Peacock, D.A. Ritchie, and G.A.C. Jones, J. Phys. C **21**, L209 (1988).

[2] C.J. Muller, J.M. van Ruitenbeek, and L.J. de Jongh, Phys. Rev. Lett. **69**, 140 (1992); J.M. Krans, C.J. Muller, I.K. Yanson, Th.C.M. Govaert, R. Hesper, and J.M. van Ruitenbeek, Phys. Rev. B **48**, 14 721 (1993).

[3] J.I. Pascual, J. Méndez, J. Gómez-Herrero, A.M. Baró, N. García, and Vu Thien Binh, Phys. Rev. Lett. **71**, 1852 (1993), and references therein.

[4] N. Agraït, J.G. Rodrigo, and S. Vieira, Phys. Rev. B **47**, 12 345 (1993).

[5] L. Olesen, E. Lægsgaard, I. Stensgaard, F. Besenbacher, J. Schiøtz, P. Stoltze, K.W. Jacobsen, and J.K. Nørskov, Phys. Rev. Lett. **72**, 2251 (1994).

[6] J.M. Krans, J.M. van Ruitenbeek, V.V. Fisun, I.K. Yanson, and L.J. de Jongh, Nature **375**, 768 (1995).

[7] J.M. Krans, C.J. Muller, N. van der Post, F.R. Postma, A.P. Sutton, T.N. Todorov, and J.M. van Ruitenbeek, Phys. Rev. Lett. **74**, 2146 (1995); L. Olesen, E. Lægsgaard, I. Stensgaard, F. Besenbacher, J. Schiøtz, P. Stoltze, K.W. Jacobsen, and J.K. Nørskov, *ibid.* **74**, 2146 (1995).

[8] A.M. Bratkovsky, A.P. Sutton, and T.N. Todorov, Phys. Rev. B **52**, 5036 (1995).

[9] C.J. Muller, J.M. van Ruitenbeek, and L.J. de Jongh, Physica C **191**, 485 (1992).

[10] T.N. Todorov and A.P. Sutton, Phys. Rev. Lett. **70**, 2138 (1993).

[11] For a perfectly cylindric neck a degeneracy is expected at $n = 2$ and 4; É.N. Bogachek, A.N. Zagoskin, and I.O. Kulik, Fiz. Nizk. Temp. **16** (1990) [Sov. J. Low Temp. Phys. **16**, 796 (1990)]. This explains the low temperature histogram on sodium in Ref. 6. The absence of degeneracy in the room temperature histograms on Au and Cu is under investigation.

[12] R. Landauer, IBM J. Res. Dev. **1**, 223 (1957).

[13] P.N. Butcher and J.A. McInnes, J. Phys. Condens. Matter **7**, 745 (1995).