Subquantum conductance steps in atom-sized contacts of the semimetal Sb

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The transition from semimetallic Sb contacts to vacuum tunnel junctions is studied using a mechanically controllable break junction. The conductance of an atom-sized contact is shown to be small compared to the quantum unit due to the large Fermi wavelength of the electrons in Sb. Observation of subquantum conductance steps when varying the contact area of atomic dimensions gives clear experimental evidence for rearrangements of atoms building up the contact.

For constrictions with diameter of the order of the Fermi wavelength of the electrons, λ_F, and for ballistic current flow the conductance is described by the Landauer formula.[1] Continuous variation of the diameter leads to discrete steps in the conductance of exactly the quantum unit $2e^2/h$, due to quantization of the transverse electron motion in the mesoscopic constriction. In experiments on a two-dimensional electron gas (2DEG) GaAs-$Al_xGa_{1-x}As$ heterostructure ($\lambda_F \approx 400$ Å) this quantum size effect has been clearly observed.[2,3] In those experiments the width of a mesoscopic constriction could be varied continuously by applying a gate voltage, reproducibly resulting in conductance plateaus at multiple integers of the quantum unit.

The mechanically controllable break junction (MCB) technique, scanning tunneling and atomic force microscopy (STM and AFM) have been used extensively in recent years to study the conductance in atom-sized *metallic* point contacts with variable constriction diameter.[4–8] Also, conductance calculations on the basis of a full dynamic simulation of metallic contact formation and fracture have been performed,[9] as well as exact calculations of the conductance through a curvilinear constriction in a 3DEG.[10] Steps in the conductance are observed experimentally as the contact size is changed by varying the pressure of the contact. However, the interpretation of the steps, which are of order $2e^2/h$ but not exactly reproducible at integer values of this quantum unit, is less clear than in the 2DEG experiments. The diameter of the metallic contact, for which the Fermi wavelength is comparable to the atomic radius, changes discontinuously due to the finite size of the atoms. Both the mechanism of atomic rearrangements[7,9] and the quantization of the conductance[8,10] were claimed to be primarily responsible for the steps.

In order to throw some more light on this important issue we present in this paper clear experimental evidence that abrupt conductance steps in Sb contacts of varying atomic dimensions arise from rearrangements of atoms in the constriction area. In analogy, abrupt conductance steps, observed in similar experiments on metals, are also believed to result from these geometrical rearrangements, irrespective of the possible additional existence of conductance quantization in these point contacts. Using a MCB atom-sized contacts are studied for Sb at liquid helium temperatures. For this semimetal a plain distinction can be made between steps resulting from the above mentioned quantum size effect and those resulting from geometrical rearrangements, due to its large Fermi wavelength.

The MCB technique is described in detail in Ref. 11. Previous MCB experiments were performed on polycrystalline metal wires. For the present experiment the samples of the very brittle single crystals of Sb are spark cut to the shape of a thin bar with approximate dimensions $0.5 \times 0.5 \times 15\,mm^3$. Into the center of the bar, which is glued on a phosphor bronze substrate, a notch to about halfway the diameter was spark cut. By bending the substrate the sample was broken at the notch at 4.2 K, under UHV conditions. These conditions ensure that the contact region between the two created surfaces, which are brought together again, is free of impurities. The size of the extremely stable contact can be adjusted on an atomic level over a wide range down to one atom by applying a voltage over a piezo element, by which the bending of the substrate is controlled. This voltage V_p is proportional to the displacement of the electrodes. From the sample geometry we estimate that 1 V corresponds to ~ 0.05–0.1 Å. The temperature for the measurements presented was between 1.3 and 4.2 K.

For metals the transition from contact to tunneling is found at about 10 kΩ. The semimetal Sb behaves quite different. The evolution of the junction resistance while scanning the transition between mechanical contact and vacuum tunneling is illustrated in Fig. 1. The current through the junction is measured at a constant bias voltage of 10 mV while the distance is controlled by the piezo voltage V_p. By decreasing V_p (smaller distance) a gradual resistance decrease, followed by a jump from ~ 4 to ~ 1 MΩ is observed. When the V_p-sweep direction is reversed after the jump, a hysteresis of about 0.1 Å in the transition occurs. The jump and its hysteresis are reproduced in detail by repetition of this approach and retreat sweep over a small V_p range. For $R > 4$ MΩ in Fig. 1 decreasing or increasing V_p over a larger range gives perfectly reproducible smooth resistance behavior, which is characteristic for the vacuum tunneling regime. At large electrode separation the resistance depends exponentially on the distance. The logarithmic slope is in good agreement with the literature value of the work function of

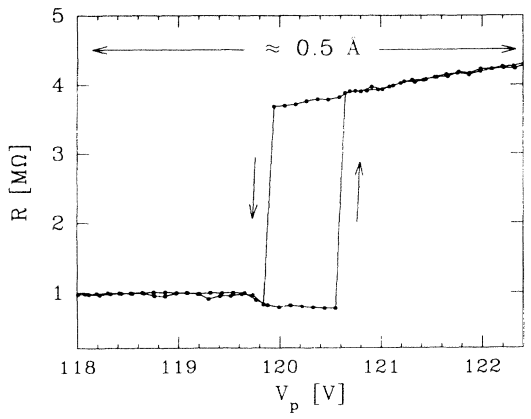

FIG. 1. The evolution of the Sb junction resistance while scanning the transition between touching and breaking of the electrodes. The vertical arrows indicate the scan directions. The measurement was performed at 4.2 K in 40 s. Note the hysteresis of approximately 0.1 Å in going from vacuum tunneling to mechanical contact and vice versa. The resistance was measured at a constant bias voltage of 10 mV.

Sb. Deviations from exponential resistance behavior observed at closer electrode separation will be described in a future publication. For $R < 1\,\mathrm{M\Omega}$ larger V_p sweeps do not reproduce: contact between the two Sb surfaces is established and the evolution of the contact area cannot be controlled reversibly.

In analogy to observations on simple metals,[5] the transition between vacuum tunneling and mechanical contact is discontinuous, with a comparable hysteresis which is as small as 0.1 Å. This jump is attributed to attractive forces between the front atoms of the two electrodes, as discussed in Ref. 5. For a metal such as Cu the resistance value just after the jump to contact, interpreted as a one-atom contact, coincides with the quantum unit 12.9 kΩ. As can be seen from Fig. 1, in the case of Sb the resistance just after the jump to contact is much larger. For a series of measurements on different Sb samples this value ranges mostly between (approximately) 1 and 2 MΩ, although values 50% larger and smaller are sometimes found.

This high resistance value can be explained by the larger λ_F of the electrons in a semimetal such as Sb compared to a simple metal. Because of the small overlap between conduction and valence band, often referred to as a pseudogap, the number of carriers per unit volume in semimetals is small compared to typical metals: in Sb $n \approx 5 \times 10^{19}\,\mathrm{cm^{-3}}$ in each band (for Cu $n \approx 9 \times 10^{22}\,\mathrm{cm^{-3}}$). In a free electron gas approximation this results in $\lambda_F \approx 55$ Å (for Cu $\lambda_F \approx 4.5$ Å). Atom-sized contacts of Sb thus have a transverse linear dimension much smaller than λ_F. This implies that in these contacts there are no ballistic conducting states at E_F, so that only tunneling contributes to the current, resulting in a conductance much smaller than $2e^2/h$.

In Fig. 2 three representative measurements of the conductance of a Sb contact as a function of V_p are shown. The traces are recorded typically in 30 s using a standard ac technique with a current modulation amplitude of a few nA. For conductance values a few times the quantum

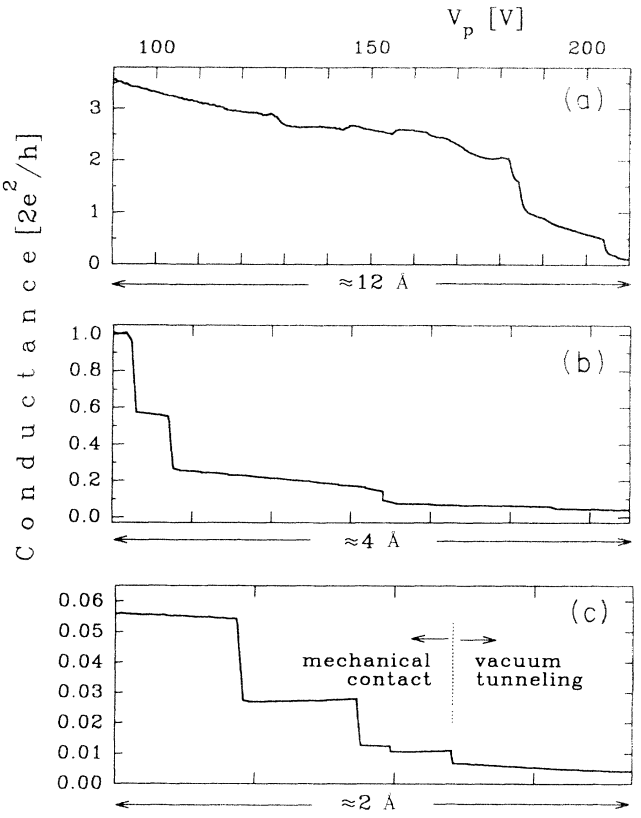

FIG. 2. Three examples of the conductance of a Sb contact at 1.3 K as a function of V_p, with V_p increasing. The three curves are recorded for three different V_p sweeps. Curve (a) shows the behavior of the conductance for large decreasing contact size. The transition from mechanical contact to vacuum tunneling is indicated in curve (c). In the mechanical contact regime shown in curve (c), as well as in curve (b), the conductance is less than the quantum unit. Conductance steps in this atom-sized contact regime are much smaller than $2e^2/h$. In all three plots every tick on the horizontal axis corresponds to 10 V over the piezo element.

unit a gradual decrease is observed with increasing V_p, as can be seen from curve (a). The absence of conductance quantization can be explained, e.g., by strong backscattering and a short length of the constriction. In curves (b) and (c) two typical curves are shown for smaller conductance values. In curve (c) the discontinuous transition between mechanical contact and vacuum tunneling is indicated. The location of this transition is without ambiguity, again because only in the vacuum tunneling regime will decreasing or increasing V_p over a larger range give perfectly reproducible smooth resistance behavior. In the mechanical contact regime shown in curve (c), as well as in curve (b), the conductance is less than the quantum unit. Similar nonquantized conductance discontinuities as in metals are observed, but in this case the size of the steps is much smaller than the quantum unit. A possible explanation of the steps in terms of conductance quantization can therefore be ruled out. The conductance steps are attributed to strain-induced atomic rearrangements in the contact, such as those found in molecular

dynamics simulations.[9,12] Unlike the 2DEG experiment, a contact area of atomic dimensions cannot be varied continuously, but at best atom-by-atom. Removing an atom from a *metallic* contact was found to result in a conductance decrease of order $2e^2/h$. Atomic variations in a *semimetallic* contact area is now seen to give rise to subquantum conductance discontinuities, which are naturally interpreted as arising from the fact that the reduction of the contact reduces the tunnel area and thereby increases the length and height of the tunnel barrier.

In summary, we have studied the conductance through atom-sized semimetallic Sb contacts of varying dimensions. Subquantum conductance steps are observed in this contact regime, giving experimental evidence for discrete atomic changes in the contact area. In analogy, this strongly supports the idea that also the origin of the abrupt conductance steps, observed in metals, is purely geometrical. On the other hand, to what extent the conductance *values* in between these steps result from conductance quantization is still under investigation. Direct experimental observation of possible conductance quantization in *metallic* contacts is complicated by the steps due to the geometrical discreteness of the contact size.

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