Magnetotransport in atomic-size bismuth contacts

Hans-Fridtjof Pernau\textsuperscript{1}, Torsten Pietsch and Elke Scheer

Department of Physics, University of Konstanz, Universitätsstraße 10, 78464 Konstanz, Germany
\textsuperscript{1} Now at Fraunhofer IPM, Heidenhofstraße 8, 79110 Freiburg

E-mail: elke.scheer@uni-konstanz.de

Received 28 April 2014, revised 26 June 2014
Accepted for publication 1 July 2014
Published 29 October 2014

Abstract

We report low-temperature transport experiments on atomic-size contacts of bismuth that are fabricated using the mechanically controlled break-junction technique at low temperatures. We observe stable contacts with conductance values at fractions of one conductance quantum \( G_0 = 2e^2/h \), as is expected for systems with long Fermi wavelength. We defer two preferred conductance scales: the lower one is in the order of 0.015 \( G_0 \) and can be attributed to single-atom Bi contact, while the higher one amounts to 0.15 \( G_0 \), as indicated by the appearance of multiples of this value in the conductance histogram. Rich magneto-transport behaviour with significant changes in the magneto-conductance is found in the whole conductance range. Although for the pristine samples and large contacts with \( G > 5 G_0 \), indications for Shubnikov-de Haas oscillations are present, the smallest contacts show pronounced conductance fluctuations that decay rapidly when a magnetic field is applied. Moreover, large variations are observed when a finite bias voltage is applied. These findings are interpreted as the transition from the diffusive to the ballistic and the ultra-quantum regime when lowering the contact size.

Keywords: bismuth, nanocontacts, atomic contacts, break junction, universal conductance fluctuations (UCF), Shubnikov-de Haas oscillations

(Some figures may appear in colour only in the online journal)

1. Introduction

Metallic atomic contacts have been intensively studied during the last 20 years, not least because they lend themselves to exploring fundamental concepts of nanophysics and quantum transport [1]. Their transport properties are described by the Landauer formula \( G = G_0 \sum T_n \) with the transmission coefficients \( T_n \) of the conduction channels, the conductance quantum \( G_0 = 2e^2/h \), where \( e \) is the elementary charge and \( h \) is Planck’s constant [2]. As a rule of thumb, a single atom contact of a metal accommodates a number of conduction channels that correspond to its chemical valence [3] with transmission coefficients given by the local band structure and the atomic arrangement. The transmission coefficients have been found to saturate at the maximum value \( T_n = 1 \), in particular for monovalent metals [1]. As a result the conductance of atomic-size metal contacts may show discrete changes of the conductance in multiples of \( G_0 \) if atoms are added to the cross section of the contact.

Because both the atomic distance in a crystal and the Fermi wavelength \( \lambda_F \) of metals amount to a few Ångströms, a nanowire as thin as one atom may act as an electron waveguide with excellent transmission properties. The nanowire is expected to show a conductance described by the Sharvin formula for a cylindrical contact, which in its simplest form, reads \( G_{Sh} = G_0 (k_F R/2)^2 \sim G_0 \), where \( R \) is the radius of the nanowire and \( k_F \) is the Fermi wave vector [1]. However, due to the coincidence of atomic size and Fermi wavelength, structural effects may conceal the waveguide properties of atomic contacts [1] because changing the diameter of the wire by one atom also changes the number of well-transmitted modes by one or a few. Consequently, atomic-size contacts and nanowires of the elementary semimetals are of particular interest
because their Fermi wavelength $\lambda_F \cong 10–50\text{nm}$ exceeds the atomic size significantly [4]. In these materials, cross-section changes of one atom should give rise to conductance steps that are much smaller than $G_0$, and do not necessarily change the number of transmitting channels. A single-atom contact of a semimetal is expected to show a conductance much smaller than $G_0$, which scales with the ratio between contact size and $\lambda_F$ as described by the Sharvin formula. Although single-atom contacts of Sb reveal a conductance of $G_{\text{Sb}} \sim 0.01\ G_0$ in good agreement with this expectation [5], the situation for Bi is less clear. In some experiments, quantized steps with amplitude $G_0$ have been found [6], while others revealed sub-quantum steps [7] or both quantized steps and sub-quantum steps [8, 9]. There is no apparent correlation between these findings and the experimental methods, which include scanning tunneling microscopy-based techniques [6, 8, 9], mechanically controlled break-junctions [5], or electromigration [7]. The appearance of quantized conduction steps has recently been interpreted as the formation of a topologically protected surface state in Bi bilayers when a nanowire is stretched to breakage [9].

However, most studies of atomic-size structures reported so far have been limited to measurement of the linear conductance or a qualitative treatment of the current-voltage characteristics [6]. This is even more surprising, as Bi in both bulk and nanostructures is known to show a large diversity of interesting transport-related phenomena because of the peculiar electronic properties of this element. Due to a slight lattice distortion, bulk bismuth is a semimetal with a very small band overlap between the valence band and the conduction band in the range of 40 meV at 0 K [10–12]. In the bulk, the electron density $n$ is strongly temperature dependent and amounts to $n = 3 \times 10^{17}\ \text{cm}^{-3}$ at 4 K, which is four to five orders of magnitude lower than for ordinary metals. The electrical conductivity of Bi nanostructures sometimes shows non-monotonic temperature dependence because of a limitation of the mean free path by temperature-independent boundary scattering [13, 14]. The Fermi surface of Bi consists of ellipsoidal pockets for electrons and holes, resulting in effective masses $m_{\text{eff}}$ ranging from 0.005$m$ to 1.2$m$ for the electrons where $m$ is the free electron mass, and from 0.006$m$ to 0.7$m$ for the holes [10–12, 15, 16]. Consequently, the Fermi wavelength depends on the band and varies between $\lambda_F \cong 14\text{nm}$ and 70 nm. In any cases, $\lambda_F$ is much longer than the diameter of a Bi atom in the lattice $d = 0.37\text{nm}$. As a result of the peculiar geometry of the Fermi surface, nanostructures of Bi show a variety of different electronic properties sensitively depending on the size, the shape and the atomic order of the structure [7, 17]. Recently large band-gap fluctuations were observed in Bi nanoparticles by STM measurements [21], pointing towards a close connection between structural and electronic properties. Furthermore, a metal-semiconductor transition and an enhanced thermo-electric figure of merit of Bi nanowires were observed [13–15]. Moreover, granular films of Bi have revealed superconductivity at temperatures as high as 6 K [22].

In the present paper we report magneto-conductance (MC) measurements of atomic-size contacts of Bi with diameters ranging from a single atom up to several atoms. The contacts were produced by stretching a nanostructured thin film of Bi to the breaking point at very low temperatures in cryogenic vacuum, and by carefully closing the gap between the two electrodes again. Our experiments reveal the rich electronic behaviour for which Bi is known [23], including conductance fluctuations [25, 26], Shubnikov-de Haas oscillations [27–29], and quantization effects [1].

2. Experimental

We use the standard techniques of electron-beam lithography, metal evaporation and lift-off to pattern a bismuth stripe (thickness 600 nm) featuring a constriction [30, 31]. By design, the width of the constriction varies linearly from 1 $\mu$m down to 100 nm. As substrate we use a gently polished bronze plate covered by a 2 $\mu$m thick polyimide layer. Note that in contrast to standard MCBJs the samples are not suspended, because the etching procedure was detrimental to the conductivity of the films. However, studies on non-suspended MCBJs of ferromagnetic metals have been performed before and reproducibly demonstrated the formation of stable atomic-size contacts [32]. A scanning micrograph of a sample is shown in figure 1(a). The thickness of the leads is 600 nm, which is reduced to about 200–300 nm in the constriction region. This size reduction is a consequence of a deposition of the material on top of the mask that reduces its width and eventually pinches it off. Thanks to this self-controlled process the mechanical force, upon bending the sample, concentrates on the constricted area and facilitates stretching the nanowire. We performed x-ray diffraction studies to reveal the type of film growth and the crystal quality; see figure 1(c). The two pronounced peaks at $2\Theta = 22.5^\circ$ and 45.8$^\circ$ correspond to the (111) and (222) directions.
of the nearly cubic structure. It is worth noting that the hole states of the Fermi surface are located in the (111) direction. From the width of the peaks and rocking curves, we determine a correlation length of 90 nm pointing to a columnar texture, as is also apparent in the SEM micrograph along the edge of a slab cut by focused ion beam; see figure 1(b). We report here transport measurements recorded on two nominally identical samples. The samples are mounted in a custom-made, three-point bending mechanism [31, 33] that is thermally anchored to the mixing chamber of a dilution refrigerator.

Conductance and MC measurements are performed at low temperatures (30 mK < \( T < 1 \) K) in magnetic fields up to 9 T and perpendicular to the sample plane. In order to measure the conductance properties with high resolution in all conductance ranges from a few hundred \( \Omega \) to the M\( \Omega \) regime, the sample is connected in series to a calibrated reference resistor of 56 k\( \Omega \), which features a very small magnetic-field dependence. The measurement circuit is voltage biased, and the voltage drop across reference resistance and sample are recorded separately to calculate the sample conductance. The transport measurements were performed using a standard lock-in electronic setup with a heavily RF-filtered measurement line in the cryostat. In order to measure the absolute and differential conductance simultaneously, a dc signal was added to the ac modulation of the lock-in amplifier. The AC voltage used for lock-in measurements at the sample was limited to below 3 \( \mu \)V, corresponding to a thermal broadening of 30 mK [33].

3. Results

3.1. Preferred conductance values

The room-temperature resistance of the pristine Bi samples is in the order of 1 k\( \Omega \) and may slightly increase or decrease towards low temperature; this variation is typical for Bi nanostructures [7, 17]. The breaking curves, i.e. measurements of the conductance as a function of elongation of the bridge, recorded at 4.2 K, reveal no or only very short plateaus at 1 \( G_0 \), but multiple plateaus at fractions of 1 \( G_0 \). The exact shape of the individual breaking curves varies from curve to curve as the atomic arrangement changes, but the general shape, e.g. not-very-well-defined plateaus with substructure and curvature, are reproducible [1]. This behaviour is typical for low-temperature measurements in particular, but not exclusively when contacts are formed with the help of lithographic MCBJs [1, 3, 18–20]. Examples of Bi breaking curves are given in figure 2(a), where a pronounced upward slope and curvature of the conductance on the individual plateaus are observed. This behaviour has been reported before for low-temperature measurements on Bi contacts formed in an STM, and explained by a distortion of the lattice and the formation of a metallic-like state with heavy electrons [8]. The transition between the steps is attributed to changes in the atomic configuration that alter the cross-section by one or a few atoms. In contrast to [8], where curved plateaus were found only above \( G = 0.15 \) \( G_0 \) in our samples they also occur reproducibly at conductance values as low as 0.015 \( G_0 \). Apparently, in our thin-film samples a different metallic-like state can be stabilized. We argue that this state arises from hole transport, because according to the x-ray analysis the <111> direction hosting the hole states may be aligned with the transport direction. Furthermore, the assumption of hole states naturally explains an increase of conductance within a plateau when stretching it further: the size of the hole pockets at the Fermi surface and their overlap increase when the lattice is elongated. Another example of plateaus with increasing conductance is aluminium [35], which also has hole pocket states at the Fermi surface (there located in the 3rd brillouin zone [34]). In Al, at the plateaus maximum, the saturation transmission of one
channel is reached, and it has been suggested that a resonant state develops by the formation of a dimer structure [36, 37]. Because of the small Fermi surface of Bi, such a resonance is not expected to develop fully. However, already a slight distortion of the lattice by the stretching process will gradually increase the band overlap, and thereby enhance $E_F$ and the transmission.

Regardless of the sign of the charge carriers, the observation of these curved plateaus confirms the formation of a metallic-like state, which is formed only in very clean contacts, of a few atoms’ size. The metallic character is further confirmed by mostly linear current-voltage characteristics and the observation of quantum-interference effects; see the next section. However, it is not straightforward to determine the conductance of the smallest contact from individual opening traces. We therefore calculate conductance histograms from repeated openings. Figures 2(b) and (c) show conductance histograms calculated from the same 80 opening traces with varying bin size and in different scaling. As a result of the non-suspended sample geometry, smaller bending radii are necessary, and thus the maximum number of opening-breaking repetitions is reduced because often the elastic deformation limit has to be overcome to stretch the wires to rupture. Figure 2(b) shows a linear plot for all data recorded in the range from 0 to 3.5 $G_0$ with a bin size of 0.05 $G_0$. No pronounced peaks are observable at 1 $G_0$ or its multiples. Figure 2(c) shows the same data in a logarithmic conductance scale, revealing broad maxima below 0.1 $G_0$ and several narrower ones between 0.1 and 1.2 $G_0$. The broad maxima at very low conductance arise from the curved plateaus described above. Because within a plateau the conductance may change by a factor of 2 or more, no narrow peaks can be expected. From this histogram we conclude that the smallest stable contacts feature conductance values in the order of 0.02 $G_0$. Figure 2(c) shows several peaks around 0.04 $G_0$, 0.07 $G_0$ and 0.15 $G_0$ and its even multiples. The structures at 0.04 $G_0$ and 0.07 $G_0$ are absent in histograms of other samples, and are therefore discarded here. We then consider the peak at 0.15 $G_0$ to be the lowest robust superstructure peak (besides the structures mentioned above around 0.015 $G_0$). We attribute the 0.15 $G_0$ peak and its multiples to somewhat larger, symmetric structures that form, e.g., when crystal planes slide along each other as proposed in [8] and [9]. The fact that the odd multiples are suppressed may indicate bilayer sliding, as suggested in [9]. Due to the film growth with columnar structure, the planes might not develop the topologically protected surface state with perfect transmission, but instead contribute with the smaller value 0.15 $G_0$ to the conductance. Revealing the exact mechanism and origin of these super-structure peaks needs further investigation. We note that the peak at 0.15 $G_0$ is visible in histograms recorded on several samples, although not in all. An alternative interpretation of the data could thus be that the fundamental peak is at 0.3 $G_0$ and all multiples...
Figure 4. Magneto-conductance (MC) and conductance fluctuation analysis. (a) MC curve of a contact measured at \( T = 150 \text{ mK} \). It reveals a bell-shape background that is subtracted by a moving averager (red line). (b) MC after subtraction of background. (c) Symmetrized signal revealing the high signal to noise ratio. For positive/negative field direction symmetric/antisymmetric part of the signal \((dG(B) \pm dG(-B))/2\) is plotted. (d) Anti-autocorrelation function of the data shown in (b). The fluctuation amplitude \( G_{\text{rms}} \) and the correlation field \( B_c \) are determined from the maximum of the AACF at zero field difference: \( G_{\text{rms}} = (\text{AACF}(\Delta B = 0))^{1/2} \) and \( B_c = \text{HFWM} \) (half width at half maximum).

would occur. On the basis of our data we cannot distinguish between these two scenarios. Below, in section 4, we will show that the magnetic field and bias dependence of the transport changes characteristically around 0.2 \( G_0 \), in agreement with both possibilities.

3.2. Shubnikov-de Haas oscillations

Because of the small Fermi surface of Bi, moderate magnetic fields are sufficient to arrive at the regime where the Fermi body is filled with a small number of Landau levels only [28]. There is a general agreement that for bulk crystalline samples, lower fields are sufficient to arrive at the regime where the Fermi surface perpendicular to the applied field, this signals a reorientation or reshaping of the Fermi surface at around 4 T. This transition becomes even more apparent when plotting \( dG \) as a function of \( B \); see figure 3(d), where we show two subsequent sweeps recorded at 4.2 K, one for increasing and one for decreasing field. A periodic variation of the conductance as a function of \( B \) would indicate a physical phenomenon, e.g., the Aharonov Bohm effect, which is related to real space rather than in \( k \) space. Also in this scaling, the peaks are not equidistant and one observes a transition from a longer period at low field \((|B| < 3.5 \text{ T})\) via a region were two periods seem to be superimposed \((3.5 \text{ T} < |B| < 5.5 \text{ T})\) to a shorter period \((|B| > 5.5 \text{ T})\). Although we cannot definitely reveal the physical origin of the periodicity changes, they suggest that the magnetic field, in combination with the low dimension, initiates a reversible structural change in the constriction that gives rise to significant changes in the charge transport behaviour. This interpretation is supported by the observation of a pronounced change in the CF in a similar field range, as we will show in the next section. Although this is a rather unlikely scenario for conventional metals, the extreme sensitivity of the electronic properties of Bi to minor geometrical changes may make this a possible scenario for Bi [50, 51].

3.3. Conductance fluctuations

We now turn to the description of the magneto-conductance (MC) measurements recorded for selected contacts. As an example, figure 4(a) shows the differential conductance \( dI/dV \) versus \( B \) of a contact with \( G(B = 0 \text{ T}) = 1.38 G_0 \) recorded at \( T = 150 \text{ mK} \). It shows a bell shape with a plateau region between \(-1.5 \text{ T} \) and 1.5 T and a saturation above \(|B| = 7 \text{ T} \) at \( G = 0.76 G_0 \). The red
Figure 5. Differential conductance as a function of magnetic field after subtraction of the background for several contacts of the same sample measured at $T < 150$ mK, revealing aperiodic, but reproducible conductance fluctuations. The traces are offset for clarity. The zero-bias conductance $dG(0)$ for each trace is indicated in the legend. The amplitudes of the two lowest traces (for the smallest $dG(0)$) have been multiplied by 10.

The remaining anti-symmetric signal shows the characteristics of white noise and reflects the high signal-to-noise ratio of the measurement with a voltage noise that amounts to $\sim 1$ nV. The bell-shaped background MC is subtracted and the traces are offset for clarity; the zero-bias conductance (ZBC) values for each trace are indicated in the legend. The most prominent observations are as follows: for high conductance there is a pronounced peak around zero-field that diminishes with decreasing conductance and is indistinguishable from the CFs below ZBC of 0.2 $G_0$. This value is somewhat smaller than the findings by Rudolph and Heremans [42] on Bi wires patterned from thin films, who observed a width-dependence of $l_p$. We note that our data could not be described with the 2D formula (as was used in [42]), which indicates that in our experiment $l_p$ was limited by the sample geometry rather than by intrinsic length scales. Also, the disappearance of the antilocalization peak for the contacts with very small conductance hints towards geometric limited transport phenomena. In this sense the obtained value for $l_p$ represents a lower limit. This analysis also indicates a lower diffusion coefficient in our columnar films than the smooth films investigated in [42].

Figure 5 summarizes typical MC curves recorded when stretching a sample starting at roughly 5 $G_0$ down to 0.014 $G_0$. The bell-shaped background MC is subtracted and the traces are offset for clarity; the zero-bias conductance (ZBC) values for each trace are indicated in the legend. The most prominent observations are as follows: for high conductance there is a pronounced peak around zero-field that diminishes with decreasing conductance and is indistinguishable from the CFs below ZBC of 0.2 $G_0$. We attribute this peak to weak antilocalization, as is expected for elements with high atomic numbers and therefore high spin–orbit coupling [39–41]. The analysis of the fitting to the 1D theory [40, 41] yields a rough estimate for the phase coherence length $l_p = 150$ nm for contacts above 1 $G_0$. This value is somewhat smaller than the findings by Rudolph and Heremans [42] on Bi wires patterned from thin films, who observed a width-dependence of $l_p$. We note that our data could not be described with the 2D formula (as was used in [42]), which indicates that in our experiment $l_p$ was limited by the sample geometry rather than by intrinsic length scales. Also, the disappearance of the antilocalization peak for the contacts with very small conductance hints towards geometric limited transport phenomena. In this sense the obtained value for $l_p$ represents a lower limit. This analysis also indicates a lower diffusion coefficient in our columnar films than the smooth films investigated in [42].

For the analysis of the CF the zero-field peak is cutout of the calculation of the AACF because the integration range is normalized accordingly to obtain the correct CF amplitude. The amplitude of the CF diminishes with decreasing $G$, so the lowest two traces in figure 5 have been multiplied by 10 for better visibility. We also note that for contacts above 1 $G_0$, the fluctuation amplitude is considerably reduced for magnetic...
Figure 7. Differential conductance $dI/dV$ as a function of applied magnetic field and bias voltage for a selection of eight contacts of the same sample, measured at low temperature $T < 150$ mK. The conductance at zero field and zero bias ($dI/dV(B = 0, V = 0)$) in the left panels are from top to bottom: 0.013 $G_0$, 0.03 $G_0$, 0.3 $G_0$, 0.65 $G_0$ and in the right panels from top to bottom: 0.9 $G_0$, 1.2 $G_0$, 1.6 $G_0$, and 2.8 $G_0$. 
fields above $|B| > 3$ T. This indicates a change in the trajectories that contribute to the interference pattern. In combination with the results from the SdH oscillations described above, we suggest a change of the transport mechanism above this field range. For the further analysis we calculated the anti-correlation functions of all CF traces and determined the rms amplitude $G_{\text{rms}}$ and the correlation field $B_c$ from its main maximum, as described above [25, 26]. Figure 6 shows the results of 35 contacts investigated in the same sample, at similar temperature $T < 150$ mK. The amplitude $G_{\text{rms}}$ increases steadily from very low values $G_{\text{rms}} < \approx 0.001 G_0$ at low conductivity and levels off to roughly $G_{\text{rms}} \sim 0.025 G_0$ at around 3 $G_0$, with rather large standard deviation. Also, this saturation value is one order of magnitude smaller than expected for universal conductance fluctuations in the diffusive regime [25, 41]. A reduction of the CF was reported for ballistic metal contacts [43], and is explained by a reduced probability of the charge carriers being scattered back through the constriction and contributing to the interference pattern $G_{\text{rms}}/G_{\text{UCF}} \sim l_\text{la}$. For Bi, with its very long $\lambda_e$ and its medium elastic mean free path $l$, this ratio is very small, and an even smaller amplitude $G_{\text{rms}}$ could be expected here. The same reduced backscattering probability may explain the reduced antilocalization peak for the smallest contacts. Similarly, it has been shown for atomic contacts of noble metals that the CF amplitude as a function of bias is sensitive with the interference of electronic trajectories being backscattered from the atomic contact and those being backscattered from the reservoir [44]. This means the CFs of atomic-size contacts are sensitive to the local structure at the contact region itself. When the transmission probability of the contact is small, the fluctuation amplitude is strongly reduced. A quantitative comparison between the model of [44] and our experiment cannot be made because of the complex Fermi surface of Bi.

The correlation field $B_c$ shows no discernible trend. Within the numerical uncertainty range it is constant $<B_c> \approx 90$ mT $\pm 10$ mT. $B_c$ describes the typical averaged ‘period’ of the aperiodic fluctuations. It is a measure for the length of the electronic paths that contribute to the fluctuation pattern. When the sample is a 2D plane, $B_c$ is translated into an effective path length by $l_{2D} = (\hbar e B_c) / w$. For wire-shaped samples with width $w$ smaller than the effective length, it has to be calculated according to: $l_{1D} = h/e B_c$ [41]. While in ballistic metal contacts, the phase area contributing to the CF is given by the temperature-independent elastic mean free path $l_e$ [43]; in the diffusive limit it is given by the minimum of either the thermal length $l_T = (\hbar D k_{\text{B}} T)^{1/2}$ or the phase coherence length $l_p$. [25, 26, 41]. However, this has only been worked out quantitatively for systems in which $\lambda_e$ is much shorter than $l$ and which, in turn, is shorter than $l_p$. Since for our samples the 2D analysis yields $l_{2D} = 220$ nm $> w = 120$ nm, the 2D approximation is not valid and we have to apply the 1D formula, from which we obtain $l_{1D} \approx 480$ nm. This value is higher than the lower estimate for $l_p$ that we obtained from the weak localization analysis above, but larger than the crystal grain structure and the film thickness in the contact area that limit the elastic mean free path. The physical meaning of this length can be revealed by temperature-dependent measurements. Up to our highest measurement temperature of 1 K, the fluctuation amplitude, as well as $B_c$, showed only a very weak variation (not shown). We therefore assume that temperature averaging is not the limiting quantity in our experiment. We conclude that $l_{1D}$ has to be identified with $l_p$, which determines the dominating interference path of the CFs as for diffusive wires. However, because the theory of CFs has not been developed for the unusual combination of length scales and because of the limited temperature range of our cryostat, we cannot certainly conclude on the physical meaning of the effective length.

3.4. Bias-dependent measurements

To get further inside the transport mechanism, we performed bias- and magnetic field-dependent measurements of the differential conductance. Figure 7 shows typical $dl/dV(B, V)$ maps of eight contacts ranging from $dl/dV(0,0) = 0.013 G_0$ to 3.022 $G_0$. Selected traces from the two lowermost panels are shown in figure 8. Pronounced variations of $G$ are observed as function of the bias and as function of the magnetic field. They are reproducible when repeating the bias or field sweep, respectively. The CFs are not symmetric with respect to bias inversion in agreement with theory and earlier observations [25, 38, 41, 40]. In addition, we observe the excitation of temporal fluctuations at higher bias levels that give rise to non-reproducible traces and show up in figure 8 as an increased noise level. To distinguish these three effects, we denote the fluctuations as function of bias as CF$_V$, the fluctuations as function of magnetic field as CF$_B$, and the temporal fluctuations as CF$_T$.

The amplitude of the CF$_V$ is reduced when applying higher bias or higher magnetic fields. The field scales at which the reduction becomes apparent depends on the contact size and the applied field. The reduction with magnetic field is in agreement with the observations described above (figure 5). It indicates a loss of phase coherence or reduction of interference path with increasing field, while the reduction of the CF$_V$ amplitude with increasing bias indicates loss of phase coherence due to smearing of the Fermi edge [40]. Alternative explanations, such as inelastic excitations of phonons [52], can be excluded because these would give rise to symmetric features with respect to bias reversal.

The CF$_T$ signal sets in at higher bias level when the CF$_V$ signal is already reduced. We attribute these fluctuations to atomic rearrangements caused by joule heating in the constriction area, although the current density is still relatively low compared to typical current densities known to exert atomic fluctuations [45–47]. This interpretation is supported by the occasional occurrence of conductance jumps (see the four topmost curves in figure 8(b)), as has been found for single-atom junctions [37]. In most cases, however, the CF$_V$ amplitudes are small and occur around a constant mean conductance value. This means that the atomic motion seems to be limited to small variations around their stable positions. Exceptions are given by the curves recorded at $B = 1$ T and $B = 3$ T in figure 8(b), where small, irreversible changes are observed. Due to the particular sensitivity of the electronic properties of Bi to small changes of the crystal structure, spatially small variations might also give rise to pronounced changes of the conductance [48–51]. Both the CF$_V$ and the CF$_T$ amplitudes
are reduced above around $|B| = 4$ T, indicating that the origin of the CF is also field dependent. In other contacts (not shown here), the CF amplitude may increase with increasing magnetic field. Such interplay between bias, magnetic field and two-level fluctuations has been observed in mesoscopic Bi samples [49, 51] and was interpreted as originating from field-dependent local double-well potentials created by structural defects. In the samples investigated here, the CF do not show a clear two-level structure, presumably because several fluctuators are active. Summarizing this part, we argue that the temporal fluctuations are caused by changes of the electronic interference paths because of dissipation-induced small-scale motion of atoms. They are modulated by the magnetic field because the field affects the phase of the electronic waves. We now discuss the dependence of the CFs on the zero-field conductance. Although for small conductance $G < 0.2 G_0$ the magnetic field dependence $CF_B$ is much smaller than the bias dependence $CF_V$, it is opposite for larger conductance. An intermediate regime where field- and bias dependence are of comparable order of magnitude is found in a range from 0.15 to 0.8 $G_0$. The suppression of the magnetic field dependence $CF_B$ for small contacts is in agreement with a transition from diffusive to ballistic transport: The origin of the pronounced negative MC (i.e. positive magnetoresistance) is the orbital magnetoresistance-i.e. the effective reduction of the mean free path, when the paths of the charge carriers are deflected by the Lorentz force [53]. When the lateral size of a wire becomes comparable to or smaller than the elastic mean free path, this effect is suppressed [54].

4. Conclusions

Summarizing, we report a comprehensive study of the magneto-conductance of atomic-size Bi contacts fabricated by the mechanically controlled break-junction technique. The transport behaviour changes twice, at around 0.015 $G_0$ and at 0.15 $G_0$. We observe conductance fluctuations that the charge transport is phase coherently over a length scale of 100 nm or more. Furthermore, we find indications for structural changes when the magnetic field exceeds a few Tesla. This structural transition change may go along with a change in the dominating carrier species-i.e. from hole to electron transport. Further measurements, e.g. of thermo power or Hall effect in strained thin films, would be required to clarify this issue.

Acknowledgements

We thank C Debuschewitz, C Schirm, V Kunej and C Bacca for valuable and fruitful discussions about this work and for their contributions in the early state of the experiments. We are grateful to A Liebig for performing the x-ray diffractometry. We acknowledge experimental and technical assistance of A Fischer. We gratefully acknowledge financial support from the Krupp foundation and from the Baden-Württemberg-Stiftung in the framework of the Research Network Functional Nanostructures.

Author Contributions

HFP fabricated the samples constructed the experimental set-up and performed the measurements. ES initiated the project, and ES and TP wrote the manuscript. All authors analyzed the data and discussed the results. All authors have given approval to the final version of the manuscript.
Funding Sources

The project was financially supported by the Alfried-Krupp von Bohlen und Halbach foundation and by the Baden-Württemberg foundation.

References

[1] Agrait N, Levy Yeyati A and van Ruitenbeek J M 2003 Phys. Rep. 377 81
[2] Landauer R 1957 IBM J. Res. Dev. 1 223
[3] Scheer E, Agrait N, Cuevas J C, Levy Yeyati A, Ludoph B, Martin-Rodero A, Rubio Bollinger G, van Ruitenbeek J M and Urbina C 1998 Nature 394 154
[4] Pippard A B and Chambers R G 1952 Proc. Phys. Soc. A 65 955
[5] Krans J M and van Ruitenbeek J M 1994 Phys. Rev. B 50 17659
[6] Costa-Krämer J L, Garcia N and Olin H 1997 Phys. Rev. Lett. 78 4990
[7] Sangioia S, Marcano N, Fan J, Morello’n L, Ibarra M R and de Teresa J M 2011 Europhys. Lett. 95 37002
[8] Rodrigo J G, Garcia-Martin A A, Saenz J J and Vieira S 2002 Phys. Rev. B. 88 246801
[9] Sabater C, Gosa’ibez-Martinez D, Fernandez-Rossier J, Rodrigo J G, Untiedt C and Palacios J J 2012 Phys. Rev. Lett. 110 176802
[10] Lax B 1956 Rev. Mod. Phys. 30 122
[11] Brown R N, Mavroides J G and Lax B 1963 Phys. Rev. 129 2055
[12] Golin S 1968 Phys. Rev. 176 830
[13] Heremans J, Thrush C M, Lin Y M, Cronin S, Zhang Z, Dresselhaus M S and Mansfield J F 2000 Phys. Rev. B 61 2921
[14] Chiu P and Shih I 2004 Nanotechnology 15 1489
[15] Cornelius T W, Brötz J, Chantko D, Miehe G, Neumann R, Toimil-Molares M E 2005 Nanotechnology 16 S246
[16] Asi C R and Hoechst H 2001 Phys. Rev. Lett. 87 177602
[17] Kaiser Ch, Weiss G, Cornelius T W, Toimil-Molares M E and Neumann R 2009 J. Phys.: Condens. Matter 21 205310
[18] Scheer E, Belzig W, Naveh Y, Devoret M H, Esteve D and Urbina C 2001 Phys. Rev. Lett. 86 284
[19] Yanson A I 2000 Atomic chains and electronic shells: quantum mechanisms for the formation of nanowires PhD thesis University of Leiden
[20] Scheer E, Konrad P, Bacca C, Mayer-Gindner A, Löhneysen H V, Hufner M and Cuevas J C 2006 Phys. Rev. B 74 205403
[21] Marchak D, Gloszmann D, Vinshtein Y, Sarby S, Lereah Y, Cheshnovsky O and Selzer Y 2013 J. Phys. Chem. C 117 22218
[22] Hamada T, Yamakawa K and Fujita F E 1981 J. Phys. F 11 657
[23] Behnia K, Balica L and Kopelevich Y 2007 Science 317 1729
[24] Behnia K, Masson M-A and Kopelevich Y 2007 Phys. Rev. Lett. 98 166602
[25] Lee P A and Stone A D 1985 Phys. Rev. Lett. 55 1622; Lee P A, Stone A D and Fukuyama H 1987 Phys. Rev. B 35 1039
[26] Altschuler B L, Kravtsov V E and Lerner I V 1986 Sov. Phys. JETP 63 441
[27] Shubnikov L and de Haas W J 1930 Proc. Netherlands R. Acad. Sci. 33 130
[28] Shoenberg D 1939 Proc. R. Soc. Lond. Ser. A 170 341
[29] Wosnitza J 1996 Fermi Surfaces of Low-Dimensional Organic Metals and Superconductors (Berlin: Springer)
[30] van Ruitenbeek J M, Alvarez A, Piñeiro Y, Grahnmann C, Joyce P, Devoret M H, Esteve D and Urbina C 1996 Rev. Sci. Instrum. 67 108
[31] Egle S, Bacca C, Pernau H-F, Hüfner M, Hinze D, Nowak U and Scheer E 2010 Phys. Rev. B 81 134402
[32] Gabureac M, Viret M, Ott F and Fermon C 2004 Phys. Rev. B 69 100401
[33] Pernau H F 2008 Elektronischer Transport in Wismutkontakten atomarer Größe PhD Thesis University of Konstanz
[34] Ashcroft N W and Mermin D N 1976 Solid State Physics (Fort Worth: Saunders College)
[35] Scheer E, Joyez P, Esteve D, Urbina C and Devoret M H 1997 Phys. Rev. Lett. 78 3535
[36] Cuevas J C, Levy Yeyati A and Martín-Rodero A 1998 Phys. Rev. Lett. 80 1066
[37] Schirn C, Matt M, Pauly F, Cuevas J C, Nielaba P and Scheer E 2013 Nature Nanotechnol. 8 645
[38] Washburn S and Webb R A 1992 Rep. Prog. Phys. 55 1311
[39] Altschuler B L, Khmel’nitzkii D, Larkin A I and Lee P A 1980 Phys. Rev. B 22 5142; Hikami S, Larkin A I and Nagaoka Y 1980 Prog. Theor. Phys. 63 707
[40] Altschuler B L and Aronov A G 1985 Electron-Electron Interactions in Disordered Systems, ed A L Efros and M Pollak (Amsterdam: North-Holland)
[41] Zyuzin A Ju and Spivak B Z 1991 Mesoscopic fluctuations of current density in disordered conductors Mesoscopic Phenomena in Solids, ed B L Altschuler, P A Lee and R A Webb (New York: Elsevier)
[42] Rudolph M and Heremans J J 2011 Phys. Rev. B 83 205410
[43] Holweg P A M, Kokkedee J A, Caro J, Verbruggen A H, Radelaar S, Jansen A G M, and Wyder P 1991 Phys. Rev. Lett. 67 2549
[44] Ludoph B, Devoret M H, Esteve D, Urbina C and van Ruitenbeek J M 1999 Phys. Rev. Lett. 82 1530
[45] Todorov T N, Hoekstra J and Sutton A 2001 Phys. Rev. Lett. 86 3606
[46] Lodder A 2005 Europhys. Lett. 72 774
[47] Trouwborst M L, van der Molen S J and van Wees B J 2006 B. J. Appl. Phys. 99 114316
[48] Birge N O, Golding B and Haemmerle W H 1989 Phys. Rev. Lett. 62 195
[49] Zimmerman N M, Golding B and Haemmerle W H 1991 Phys. Rev. Lett. 67 1322
[50] Birge N O, Moon J S and Hoadley D 1996 Czech. J. Phys. G 16 2343
[51] Weiss G and Brouer S 2001 Europhys. Lett. 54 654
[52] Naidyuk Yu G and Yanson I K 2005 Point Contact Spectroscopy (Berlin: Springer)
[53] Abrikosov A A and Gorkov L P 1962 Zh. Eksp. Teor. Fiz. 42 1088; 1962 Sov. Phys. JETP 15 752
[54] Scheer E, Löhneysen H V, Mirlin A, Wölfle P and Hein H 1997 Phys. Rev.Lett. 78 3362