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2007 J. Phys.: Condens. Matter 19 016212

(http://iopscience.iop.org/0953-8984/19/1/016212)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **19** (2007) 016212 (11pp) doi:10.1088/0953-8984/19/1/016212

Is there ballistic transport in metallic nano-objects? Ballistic versus diffusive contributions

N García[1], **Ming Bai**[1], **Yonghua Lu**[1], **M Muñoz**[1], **Hao Cheng**[1] **and A P Levanyuk**[2]

[1] Laboratorio de Física de Sistemas Pequeños y Nanotecnología, Consejo Superior de Investigaciones Científicas (CSIC), Madrid 28006, Spain
[2] Fisintec Innovación Tecnológica, Miraflores 65, Alcobendas, Madrid 28100, Spain

Received 30 August 2006, in final form 15 November 2006
Published 7 December 2006
Online at stacks.iop.org/JPhysCM/19/016212

Abstract

When discussing the resistance of an atomic-or nanometre-size contact we should consider both its ballistic and its diffusive contributions. But there is a contribution of the leads to the resistance of the contact as well. In this context, the geometry and the roughness of the surfaces limiting the system will contribute to the resistance, and these contributions should be added to the ideal ballistic resistance of the nanocontact. We have calculated, for metallic materials, the serial resistance of the leads arising from the roughness, and our calculations show that the ohmic resistance is as important as the ballistic resistance of the constriction. The classical resistance is a lower limit to the quantum resistance of the leads. Many examples of earlier experiments show that the mean free path of the transport electrons is of the order of the size of the contacts or the leads. This is not compatible with the idea of ballistic transport. This result may put in serious difficulties the current, existing interpretation of experimental data in metals where only small serial resistances compared with the ballistic component of the total resistance have been taken into account. The two-dimensional electron gas (2DEG) is also discussed and the serial corrections appear to be smaller than for metals. Experiments with these last systems are proposed that may reveal new interesting aspects in the physics of ballistic and diffusive transport.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

When talking about electron transport, the condition needed for having ballistic transport through an object of size D is that the mean free path of the electrons $l \gg D$. Then the electron does not suffer collisions and goes throughout the object ballistically. In contrast, if $l \ll D$, then conduction is controlled by the ohmic resistance. Figure 1 illustrates the problem.

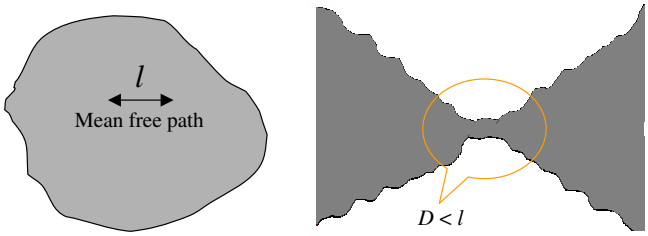

Figure 1. The geometry scheme for the relation between mean free path l and the size of the object D. When it comes to the nanoconstrictions, the size of the constriction becomes smaller than the mean free path.

But we notice already a difficulty in defining our problem, i.e. that the boundaries of the bulk material play no role because the surface area is negligible. However, when dealing with nano-objects, in our case with nanoconstrictions, we immediately have to put ourselves the question: what is now the effective mean free path of the electrons? Is it the same as for the bulk? Or, more importantly, what is the influence of the surface roughness, not only in the constriction region but also in the leads connecting the constriction?

Experimental data on conduction of narrow metallic constrictions and nanocontacts have been interpreted as evidence for conductance quantization of the electrical current as a consequence of ballistic transport. It was calculated that the conductance σ is quantized by intervals of integral fraction units of $\sigma^{-1} = R_0 = h/2e^2 \approx 12\,900\ \Omega$. The data were interpreted [1–3] using Landauer's formula [4] and the geometry in the left-hand side of figure 3(B) for $\delta > \lambda$ with the restrictive condition of $\theta = \pi/2$, where λ is the Fermi wavelength of the electrons. But no roughness, either in the leads or at the contact, was taken into account. Experiments [5–10] apparently showed that the corresponding conductance or resistance plateaus at the predicted 'quantized' values were in agreement with the theory. Histograms of conductance on a 'large' number of samples showed peaks around the integral numbers of conductance. Figure 2 presents results of two such experiments, as reported in [6–8]. However, an important point is that, in most experiments, the geometry of the metallic shape leading to 'quantization' remained unknown. As one of few exceptions, the experiments with Au of Ohnishi *et al* [9] showed the values of conductance at integer numbers and, at the same time, the geometry of the constriction observed by TEM (see figure 3). In figure 3(a), one can see how the contact is reduced and broken. Also, in figure 3(a), a more detailed picture is given with the value of the conductance measured at the same time. However, one should notice that the angle leading to the constriction (see figures 1 and 3) was not $\theta \approx \pi/2$ but $\theta \approx \pi/4$, a quite important difference, as will be revealed. In addition, as can be seen in figure 3, the Au surface was quite rough. The general view, up to now, of the problem of transport in atomic-size metallic contacts at room temperature (RT) and low temperature (LT) is mainly determined by the ballistic conductance through the narrowest part of the contact. The rest of the contributions, including the contributions of the leads, are considered as small corrections [10].

2. Theory and comparison with experiments

In this paper we would like to discuss the different contributions to the resistance of metallic nanocontacts and especially the contribution of the leads. This will help us to understand whether the previous interpretation is affected by small corrections or not. The resistance of

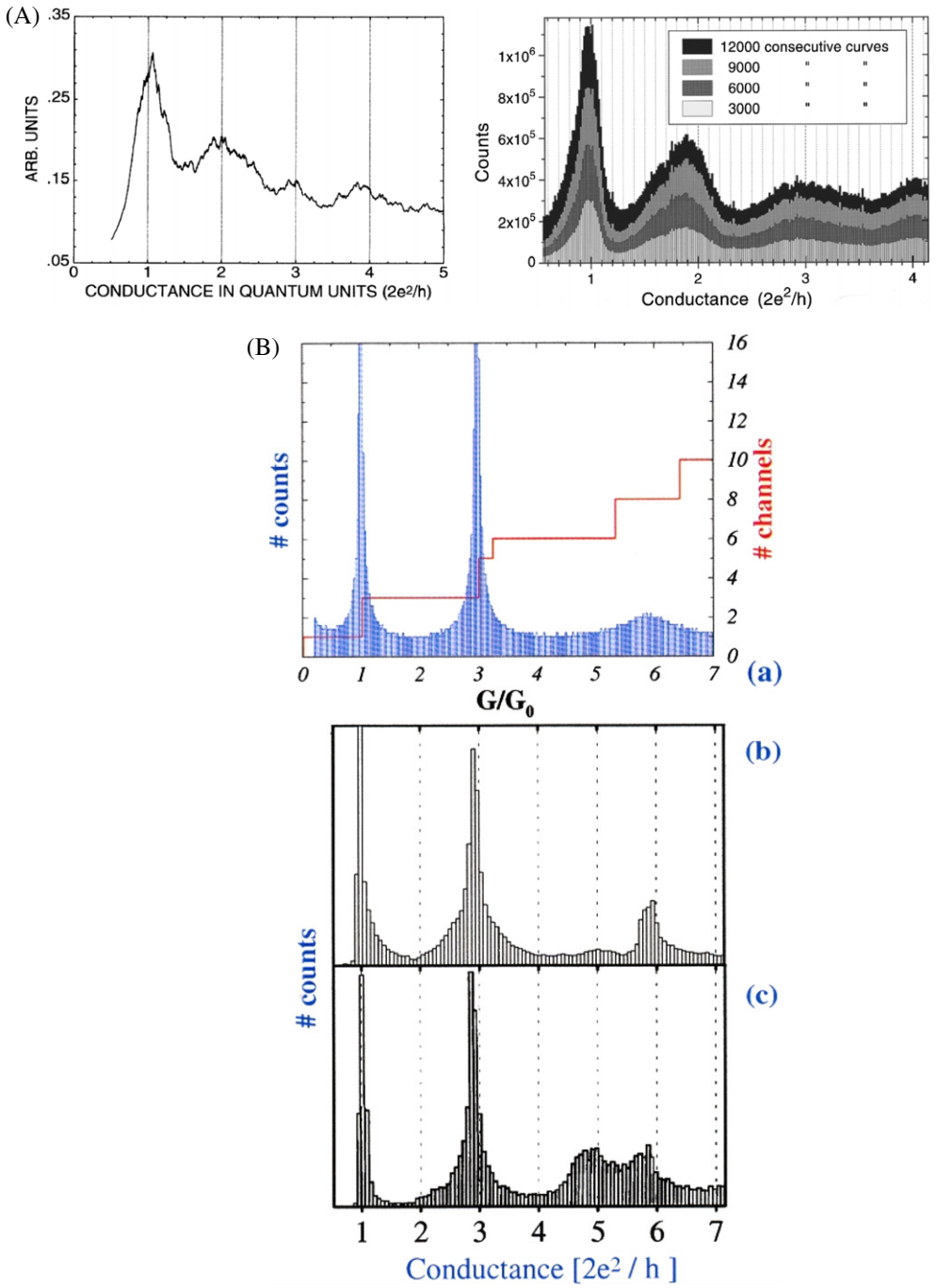

Figure 2. (A) Experimental histograms obtained from thousands of contact breakings, showing clear evidence of a preference for conductance values close to integer multiples of G. After [6] and [8]. (B) (a) Conductance histogram obtained from the average conductance G/G_0 versus displacement d/λ_F. The stepped line indicates the number of channels contributing to each conductance value. (b) Conductance histogram for Na obtained from hundreds of simulations of contact-breaking processes. (c) Experimental conductance histogram for Na. After [11], [39] and [7].

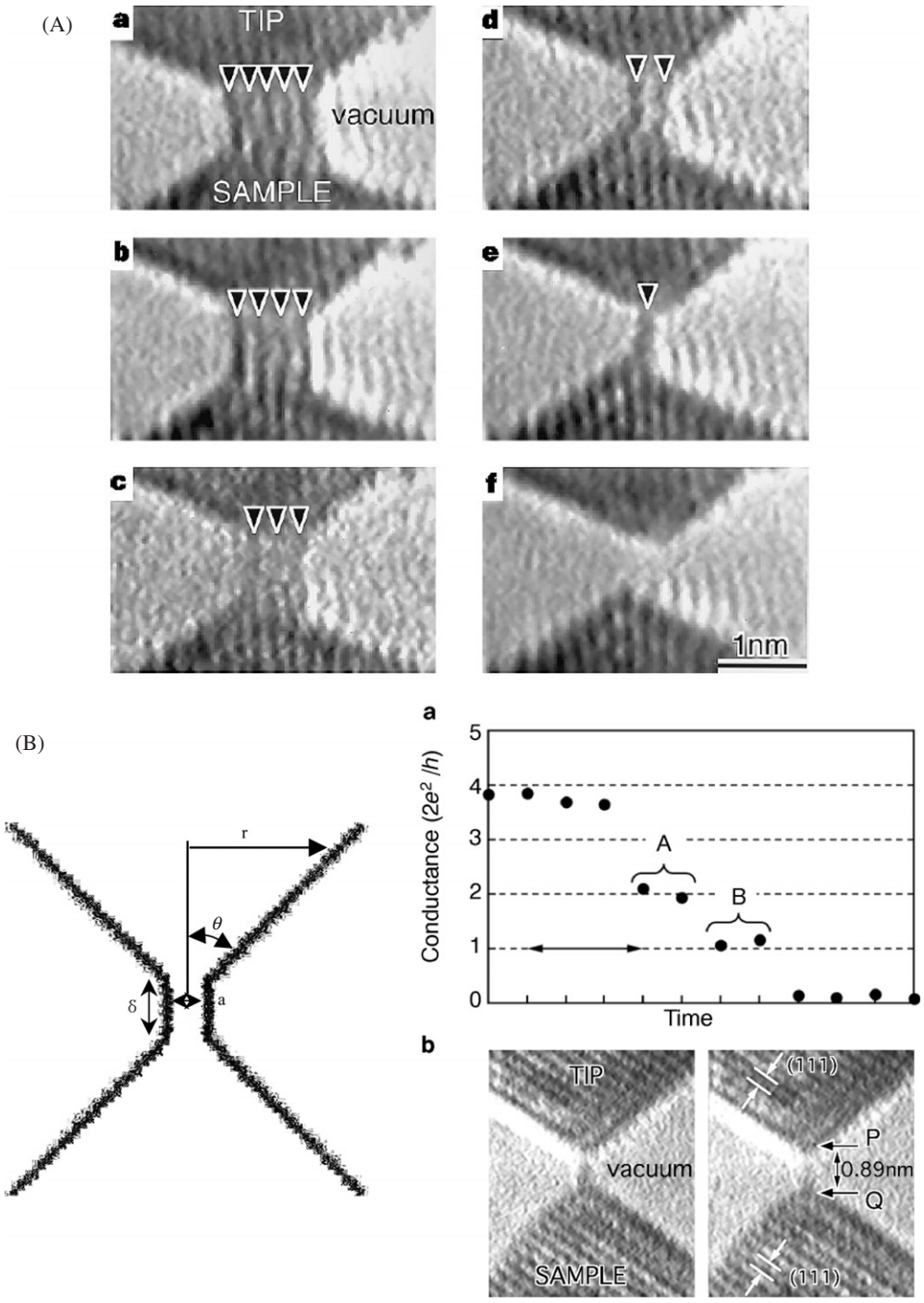

Figure 3. (A) Direct observation of the neck-breaking process of gold contacts in a STM–TEM setup. After [9]. (B) TEM images of gold contacts showing a half opening angle of approximately $\pi/4$ and the measured 'quantized' resistance for different stages of the contact. After [9]. The left is the corresponding geometry used for calculations. Profile of cone-like nanocontact geometry, where a and δ is the narrowest width and the length of contact, θ is the half open angle and r is the radius of the cone.

the 'neck' has been discussed [11–16]. It was shown that the corrections due to the roughness of the neck or from point imperfections at the neck could give appreciable corrections. However, there have been few discussions of the contributions of the leads to the resistance and, certainly, roughness has not been considered. The only considerations taken into account for these contributions were made using Wexler's work [17] to interpolate between the Sharvin–Knudsen ballistic [1] and Maxwell classical conductance for an orifice of diameter a in an isolating screen separating two conducting half spaces; i.e. a geometry with $\theta \approx \pi/2$ and $\delta \ll \lambda$. The formula reads:

$$R = R_s + \gamma(\kappa)\rho/a = R_s\left(1 + \frac{3\pi}{8}\gamma(\kappa)a/l\right) \tag{1}$$

where l is the mean free path of the conducting electrons, $\kappa = a/l$, and $\gamma(\kappa)$ is a factor of the order of unity, which is 1 when κ tends to infinity and 0.67 when κ tends to zero. The Sharvin–Knudsen resistance is given by,

$$R_s = 4\rho l/3A = \left(\frac{2e^2}{h}\frac{k_F^2 A}{4\pi}\right)^{-1} \tag{2}$$

where $A = \pi a^2$ is the area of the orifice, k_F is the Fermi wavevector of the electrons respectively, h and e are the Planck constant and the electron charge. This resistance dominates in the limit of $\kappa \ll 1$. To the opposite limit, when $\kappa \gg 1$, one has the Maxwell result ρ/a. Then the Sharvin resistance is identified as the quantum resistance. Quantum resistance jumps show up at the Sharvin background, as seems to be revealed in the experiments. It is from this formula that one concludes that, at $a \ll l$, the main contribution to the contact resistance is the ballistic resistance, because the corrections are of the order a/l. We remark that Wexler's argument is a classical one without consideration of roughness. In this case, with the given geometry ($\theta \approx \pi/2$ and $\delta \ll \lambda$), we have no objections that the estimations of the corrections are valid.

However, it is clear to us that the use of Wexler's formula is irrelevant for the real case of atomic-size metallic contacts. Wexler's formula corresponds to $\theta = \pi/2$ geometry for specular scattering in the walls and no roughness at the surface. This implies that the mean free path of the bulk material is the same as that at the vicinity of the orifice.

It is difficult to estimate the role of the roughness for $\theta = \pi/2$. However, for smaller angles of θ this effect should be very large, because the resistivities as well as the effective mean free path change due to surface scattering by the walls of the leads which contact the orifice.

In 1901, Thomson [18] first suggested that the source of the high resistivity, ρ, of very thin films of metals lies in the limitation of the mean free path of the electrons due to non-specular scattering at the surface of the films. With the advent of low temperature and thin-film growth techniques, Lovell [19] and Andrew [20] realized experiments proving the suggestion of Thomson. These experiments were performed in Sn films and Na wires. The theoretical work was done by Fuchs [21], Sondheimer [22] and Chambers [23] on thin films using the Boltzmann equation and taking into account the specularity in the p parameter for the surface scattering to explain the data. This is a qualitative parameter that, for $p = 1$, we obtain the limit of specular scattering (surfaces do not contribute to the resistance); and for $p = 0$ we have pure diffusive non-specular scattering, in which case we have a large contribution to the resistance due to the surface limiting of the system at hand. Dingle [24] and MacDonald and Sarginson [25] did the same work for circular and square section wires, respectively. These results are also described in a book on metals edited by Ziman [26]. These earlier experiments were done in wires having diameters of microns, but to discuss the ballisticity in metals one does not need to go to

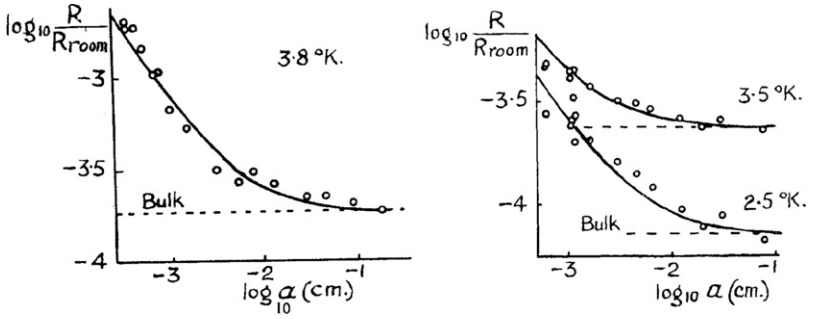

Figure 4. (Left) Resistivity of tin foils at 3.8 K against foil thickness. The full line is the theoretical curve for diffuse scattering. (Right) Resistivity of mercury wire at 3.5 and 2.5 K against wire diameter. The full line is the theoretical curve for diffuse scattering. After [20].

nanometre size, which is necessary for RT experiments. However at LT (liquid helium or so) the electron mean free path for a bulk metal of good purity may be of the order of centimetres. Therefore, a section of wires of microns may become smaller than the mean free path, and, in theory, we have ballistic transport. However, this does not happen because, as we decrease the wire section, the resistivity of the wire increases, reducing the effective mean free path, as illustrated in the earlier experiments (figure 4) [20]. Therefore the ballistic limit is not reached because the ohmic resistance keeps increasing. The same conclusions can be reached from the most recent and active field of thin films, where technology has been greatly improved [27–32].

From earlier and recent work, theory and experiments are in agreement [18–34]. We can resume the behaviour of the resistivities for different geometries (films, circular and square wires). These are as follows for the ratio $\kappa = a/l$, where a is the smallest transverse size of the object at hand (for example, the thickness for films or the diameter for wires) and l is the mean free path in the bulk [22]: For thin films:

$$\frac{\rho}{\rho_0} = \frac{4}{3}\frac{1-p}{1+p}\frac{1}{\kappa \log(1/\kappa)} \qquad \kappa \ll 1. \tag{3a}$$

For circular wires:

$$\frac{\rho}{\rho_0} = \frac{1-p}{1+p}\frac{1}{\kappa} \qquad \kappa \ll 1. \tag{3b}$$

For square wires:

$$\frac{\rho}{\rho_0} = \frac{1-p}{1+p}\frac{0.897}{\kappa} \qquad \kappa \ll 1. \tag{3c}$$

From the above classical limits, for $\kappa = a/l \ll 1$, ρ varies approximately as ρ_0/κ (where ρ_0 is the bulk resistivity due to collisions). This means that the effective mean free path for the object l_e is controlled by the transverse size a. Therefore it seems that the mean free path of the material, l, plays a smaller role than when it is smaller than a. In figure 5(a) we present a collection of experimental data justifying the above conclusion. Then in a nanosystem or nanoconstriction ($a \approx 1$ nm), the mean free path for collisions in metals is around 20–40 nm, but this plays no role in the resistance. Instead, the role is played by scattering at the rough surface characterized by l_e which is much smaller. However, this result presents serious difficulties in reaching the statement that $a \ll l_e$ is the criterion for ballistic conductance. As has been seen, it does not help to reduce the constriction width if, at the same time, the roughness is not reduced, since metals have a natural roughness that cannot be reduced below a limit.

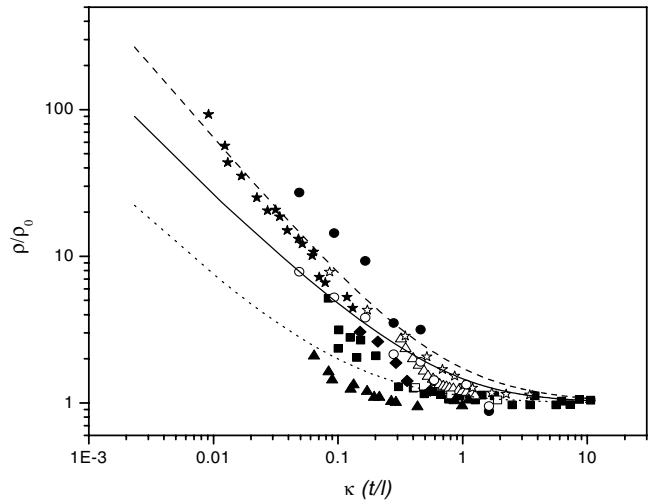

Figure 5. A summary of measured resistivities of thin films versus thickness of several metals retrieved from the corresponding references (-■- Pt [27], -●- Au ($T \approx 0$ K) [28], -○- Au ($T = 250$ K) [28], -△- Bi [29], -☆- Cu [30], -□- Au [31], -◆- Ni–Fe Permalloy [32], -★- Al [33], -▲- $CoSi_2$ [34]). The resistivities are normalized by the metal bulk resistivity ρ_0, and the thickness is scaled by free mean path l accordingly. The experimental data are fitted using the Fuchs–Sondheimer theory for the best values of p as well as the Mayadas and Shatzkes theory. The dashed line is for ρ by the M–S theory with $p = 0.7$ and $R_{MS} = 0.3$. The dotted line and the solid line is for ρ by the F–S theory with $p = 0.7$ and 0, respectively.

In addition to the surface scattering, Mayadas and Shatzkes [33] noticed that, in addition to surface scattering, grain boundary scattering in polycrystalline films will also increase the resistivity. The resistivity increases as films become thinner, because the grain boundaries reduce their size. In figure 5, we sum up the data on thin film resistivities for several metals together with data fitting lines using the Fuchs–Sondheimer theory for the best values of both p and the parameter R_{MS} of the theory for grain boundary scattering [33].

The theory has been formed using the classical Boltzmann's equation. However we may put to ourselves the question of what would happen if we performed quantum calculations. First we stress that we are not discussing the ballistic part of the resistance but the contribution of the diffusive scattering due to roughness in the leads. Second, the classical calculation results give a lower limit of resistance when compared with quantum calculations. Already, for thin Au films [34], it was shown that the resistivity by the quantum approach increases more dramatically than predicted by classical theory when the thickness is reduced. A crossover is found for ρ behaving from a^{-1} to a^{-2} [34] as the film thickness is reduced. This is important because quantum calculations are complicated and the classical approach permits us to set lower bounds on the effect. The result can also be discussed from ideas generated in the localization of electrons in disordered media. Classical arguments do not lead to localization; however, by using quantum mechanics, localization appears as disorder increases in the conductor. This implies less and less ballistic behaviour and, at the end, one has to obtain localization by reducing a; that is to say, quantum calculations will yield values of resistance higher than those calculated from classical mechanics.

To estimate the contribution of the roughness in the leads, we will start to discuss the problem with the cone-like model [35] of figure 3(b). Quantum calculations performed for this model did not take into account the roughness either in the neck or in the leads; i.e. only

Table 1. Classic calculations for ohmic resistances of the leads, taken to one-atom contact for different materials.

$\theta = \pi/4$				$\theta = \pi/6$	
R (ohms)	$p = 0.3$	$p = 0.5$	$p = 0.7$	R (ohms)	$p = 0.7$
Au	9 240	5 500	3 190	Au	4 070
Na	19 950	11 875	6 887	Na	8 787
Sn	46 200	27 500	15 950	Sn	20 350

specular scattering in the cone walls was considered. This is not very realistic for metal because, from our previous analysis, metal surfaces have significant roughness, as is confirmed by theory and experiments. Also, the scattering is non-specular, with p different to unity, in the model of Fuchs–Sondheimer for Au, Na, Pb, Pt or for any metal. Therefore a study of the value of the R_{ohm} due to the rough walls of the cone leading to the contact constriction is in order. We already advance that the ohmic resistance for the observed values of $\theta \approx \pi/4$ [9] is of the same order as for Sharvin's resistance.

To perform the calculation, we use the resistivity for a circular wire $\rho(r, p)$ as in the theory of Dingle (see [24] and [22] formulae 28–31). Figure 6(A) shows the resulting resistivities for different p values as a function of the wire radius. Notice that $\rho(r, p)$ grows with r^{-1} for small values of r. The resistance of the cone with angle θ is obtained as a superposition of cylinders of radii r and the same length given the non-locality of $\rho(r, p)$. This can reasonably be approximated as:

$$R_{\text{ohm}} = 2\int_0^{r_M} \frac{\rho(r, p)}{(\pi a^2/4 + \alpha(\theta) r^2)}\,\mathrm{d}r \tag{4}$$

where $\alpha(\theta)$ is the solid angle.

We proceed now by applying the previous estimations to the experiments by Ohnishi *et al* [9]. These represent a set of experiments where the geometry was observed at the same time that the conductance was being measured (see figure 3(B)). We notice that the experimental angle was $\theta \approx \pi/4$. These results underestimate the ohmic resistance as observed from fitting data for films in figure 5, but are nevertheless large. The R_{ohm} values obtained for a one-atom orifice $a \approx 0.3$ nm are, by substituting the values of ρ_0 as 2.2 $\mu\Omega$ cm, 4.75 $\mu\Omega$ cm and 11 $\mu\Omega$ cm for Au, Na and Sn respectively [36], of the order of 10 000 Ω and more. In table 1 we present the values for different p values. If we compare these values with those observed in experiments of quantized resistance [5–10], one will notice that they are of the same order. In addition, we would like to mention that the above result showed that the effective mean free path at the constriction is of the order of a, and R_{ohm} behaves as a^{-2}. Therefore the Sharvin and ohmic resistances are both of the same order and have an a^{-2} dependence (slope of the lines in figure 6); i.e. both resistances are indistinguishable in the classical approximation. So there is no way to distinguish in the experiments which part of the resistance is contributed by the leads and which is contributed by the constriction.

The estimations presented here call for some precautions about the interpretation of the experimental data [5–10] as resistance is contributed only by the constriction and with a ballistic character. We see that the leads may contribute as much as the constriction, and this cannot be ignored in metallic nanocontacts. Therefore, reconsideration of the interpretation of the data on small nanocontacts seems necessary. A pertinent question may be what the conditions should be for the contribution of the leads to be negligible compared with the ballistic part from the neck? We concluded that, in order to have a negligible value of R_{ohm}, one needs:

(i) that the surfaces of the walls of the leads are specular, which is difficult to achieve because

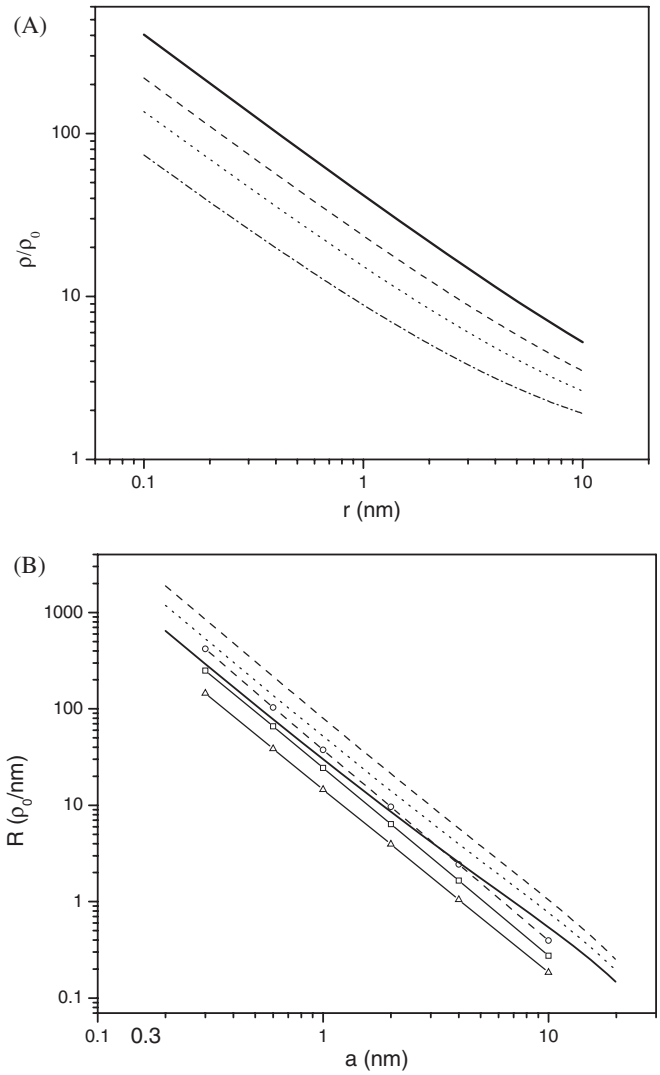

Figure 6. (A) The resistivities for different p values as a function of the wire radius, based on the analysis from the theory of Dingle [24]; see also [6]. The solid line, dashed line, dotted line and dash-dotted line represent $p = 0$, 0.3, 0.5 and 0.7, respectively. (B) The resistance of the cone integrated from a given diameter to a, with fixed $\theta = \pi/4$ for different $p = 0.3$, 0.5 and 0.7 in dashed, dotted and solid lines, respectively. The comparison by simulations are with -○-, -□- and -△- lines for $p = 0.3$, 0.5 and 0.7, respectively.

not even the best grown films have this characteristic (real metal surfaces are rough);

(ii) that the ballistic and ohmic resistances, which are practically additive for specular scattering, are strongly non-additive for non-specular scattering, therefore it seems physically unsound and hard to prove or;

(iii) that the geometry of the nanocontacts have $\theta \approx \pi/2$, which is also a peculiar condition and, in fact, the geometry we know in [5] has $\theta \approx \pi/4$ with a large ohmic resistance (see table 1).

3. The two-dimensional electron gas (2DEG)

As is well known, the first experiments showing conductance quantization were reported on work performed using a 2DEG by two groups at the same time [37, 38]. In this system, the experiments, especially the geometry, are well controlled and as many as ten plateaus in the conductance curve could be observed. In this case, it is likely that the influence of the leads is less important, not only because the geometry was more than $\theta \approx \pi/2$, but also because the surfaces should be much smoother than those of the metal constrictions. The geometry in this case is two-dimensional; the variation in resistance for the triangular geometry ending in a width W, equivalent to the conical geometry discussed here, behaves as $\log(W)$. More interestingly, the constriction is made by a gate voltage. This is important, because the scattering of the electrons by the boundaries is not against matter (a hard wall potential), as in the case of metals, but against the electrostatic potential (having parabolic-like behaviour, softer than a hard wall), which reduces the scattering strength.

The engineering of the 2DEG devices has come to a good controlled fabrication stage, and it can be used to clarify the importance of the contribution of roughness to the resistance. It would be interesting to fabricate a 2DEG with well-controlled constrictions with different geometries and roughness to compare the different conductances for the different geometries. This should be feasible, and will be a clear way to discriminate and understand better the scattering contributions to the resistance. In our opinion, this is probably the only way to discriminate between diffusive and ballistic contributions, since the condition of ballisticity is strongly affected by the geometry.

Acknowledgment

This work has been supported by the European Union's MUNDIS project.

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