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Normal Metallic Point Contacts

The (nonohmic) resistance of tiny metal contacts shows new structure at metal phonon energies.

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With the widespread use of miniaturized semiconducting devices (diodes, transistors, and integrated-circuit chips), new problems arose in attaching stable metallic leads. Although the currents that flowed through the metallic leads were small—typically of the order of a few milliamperes—the contacts were so small that the resulting current densities were enormous—as large as 10^6 amperes per square centimeter. Such high current densities were well above anything encountered in the laboratory or in technology before. In such a regime, new phenomena, which in the ordinary world are very tiny, become important. All metallic crystals contain impurities, vacancies, or dislocations. In a strong current density (or equivalently, a strong electric field), these defects force electrons to stream around them and hence exert forces on them; they move or diffuse, driven by the electron "wind." This process is now called electromigration (*1*). When such diffusion takes place in constant strong fields for a long time, the defects migrate and become trapped—for example, at the metal-semiconductor interface. Mechanically, contacts with piled-up vacancies and impurities become weaker and weaker—and eventually break. These technical problems are now well understood, but the solution has involved a wide variety of disciplines.

Summary. The measured voltage derivative of the nonlinear resistance of tiny point contacts can be separated into a phonon-emission effect (α^2F) and an analytic functional form (background effect). The α^2F's show structure coincident with bulk phonon densities of states. Values of the integral of 2 α^2F/ω are closely related to literature values. The background effect is related to the impurity concentration of the materials.

Background: Large Contacts

The general problem of understanding electrical contacts is quite old: it first arose with the advent of rotating electrical machinery in the early 1890's. The classical solution in the ohmic regime was given by Maxwell (*2*) by solving Poisson's equation for the electrical potential V as function of coordinate $\vec{r}$

$$\nabla^2 V(\vec{r}) = 0 \tag{1}$$

in oblate spherical coordinates. Contours of constant potential of current flowing through a constricting circular orifice of radius a are given by

$$V(\vec{r}) = \pm V_0[1 - \frac{2}{\pi} \arctan (1/\xi)] \tag{2}$$

where the oblate spherical coordinate ξ may be found from the more familiar spherical polar coordinates r and z by solving the implicit equation $r^2/a^2 = (1 + \xi^2)(1 - z^2/\xi^2 a^2)$. The two signs refer to the two sides of the circular orifice: the geometry has cylindrical symmetry. The resistance of such a contact may be found by dividing the total voltage drop (here $2V_0$) by the total current flowing through any contour of constant voltage. For simplicity, we choose the contour $V(\vec{r}) = 0$; that is, the orifice itself ($\xi = 0$). The current I is given (in cylindrical coordinates) by an integral of the current density $\vec{J}$ over the area $\vec{O}$ of the orifice

$$I = \int d\vec{O} \cdot \vec{J} = \int_0^{2\pi} d\varphi \int_0^a rdr\, \sigma\left(\frac{\partial V}{\partial z}\right)_{z=0} \tag{3}$$

where σ is the conductivity and $\partial V/\partial z$ is the z component of the electric field $\vec{E}$. The Maxwellian resistance (*2*) of the contact is then given by

$$R_M = \frac{\rho}{2a} \tag{4}$$

where ρ is the resistivity ($\equiv 1/\sigma$). Equation 4 has been verified (*3*) to an accuracy of about 1 percent for a wide variety of "practical" contacts. There are also many exceptions (*3*).

New Phenomena in Small Contacts

In this article we focus on some interesting exceptions that have been examined (*4, 5*) at Nijmegen. In these experiments the contact radii are made so small (~ 40 angstroms) that the current densities are orders of magnitude (10^{10} to 10^{11} amp/cm^2) larger than those previously considered. We were stimulated to try such experiments by the work of Yanson (*6*), who used a technique that depends on puncturing two metallic films, separated by a thin (100 Å) insulating layer, with a burst of voltage coupled through a high resistor. It oc-

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curred to us that if we could form such tiny metallic bridges from sharp points, the resulting junctions might be simpler to form and control than those formed from shorted films. The major potential disadvantages of point contacts involved questions of mechanical stability.

Experimental technique. We formed a sharp tip (of radius ~ ½ micrometer) on a thin wire ("spear") by an electrolytic etching technique (*7*). This was mounted rigidly on a subassembly with a larger wire ("anvil"), cooled to 1.2°K, and the spear-anvil separation was carefully adjusted. On the basis of the results discussed below, we believe that within the etched tip of the spear there are microscopic regions which, on this scale, are large conical mountains "rising" 100 to 200 Å above the average "plane" of the tip. Usually one of these is dominant, actually touches first, and forms the metallic, resistive junction we measure. The separation is first crudely adjusted by means of differential screws and then finely adjusted by using a piezoelectric substage. We can control the separation to better than 10^{-6} cm by means of the piezoelectric lever arrangement.

The experimental apparatus is shown in Fig. 1. The resistance of the point contacts formed in noble metals at a low temperature (1.2°K) is quite high—about 10 ohms. If we estimate the radius a_M of such a contact from the Maxwell equation (Eq. 4), we find the tiny value of about 0.2 Å. This is far smaller than both the classical resistive scattering length l (about 10^4 Å) and the quantum-mechanical de Broglie wavelength Λ (about 5 Å). Moreover, the measured contact resistance is distinctly nonlinear, or nonohmic.

Nonlinear contacts. In Fig. 2 we show the voltage dependence of the current and the first and second derivatives of a nonlinear I-V characteristic curve for a point contact of copper, whose resistance was 5.7 ohms at a voltage near zero. [The measuring technique involved phase-sensitive detection of first and second harmonics, similar to that used in tunnel-junction spectroscopy of superconductors (*8*).] Figure 2 is a direct photograph of the output of an x-y plotter. The (dynamical) resistance (curve b) is nearly an exactly symmetric function of voltage, the nonlinear second derivative (curve c) nearly an antisymmetric function. The broad peaks in curve c at ± 15 and ± 27 millivolts are reproducible from sample to sample and contact to contact. The small structure near zero bias and the narrow dip at +17 mV are electrical noise. The tiny ripple seen at > 30 mV is the intrinsic mechanical and electrical noise in the experiment. New phenomena have arisen because of the small size, high current density, and high electric field of the point contacts.

Scattering Length versus Radius

Let us consider the classical scattering length question first. The problem of the flow of classical electrons of large scattering length l through a small orifice resembles a problem in gas kinetic theory (*9*). As the pressure in a gas vessel is lowered, for example by pumping through a small hole into a vacuum, the mean free path l between collisions increases. Eventually the path length will be larger than the orifice radius a. One then no longer has diffusive flow, and the gas molecules penetrate the orifice ballistically. This problem was first considered by Knudsen (*10*) in 1909, and is now called the Knudsen effect. The different regimes are characterized by the Knudsen ratio $K = l/a$.

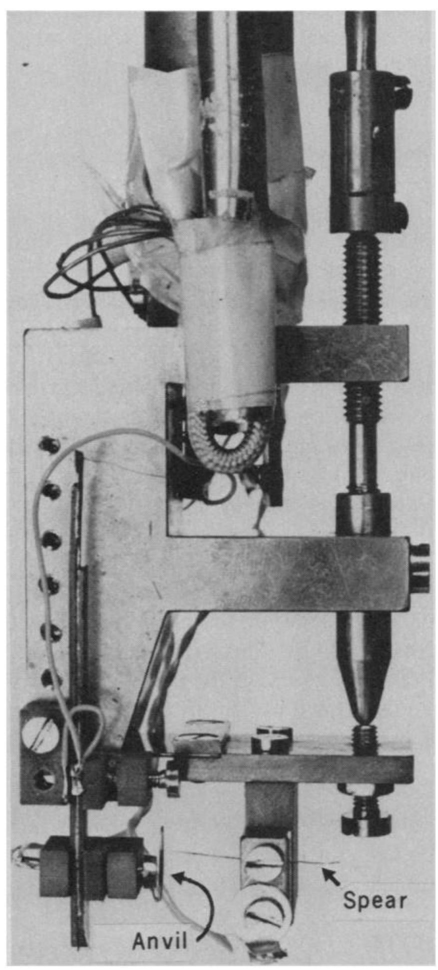


Fig. 1. Experimental apparatus. The "spear" on the right is moved into the "anvil" on the left by means of differential screws and a piezoelectric lever arrangement. The separation can be controlled to better than 10^{-6} cm.

In the case of electrical contacts, the major effect (in the regime of high Knudsen ratios) is to cause a steep voltage gradient or high effective electric field to develop in the contact region. Crudely, then, in this regime the contact serves as a device to inject electrons of "high" kinetic energy from one metal into another, but with a carefully controlled excess kinetic energy or velocity. This excess velocity is given by $\Delta v = \pm eV/p_F$, where the sign depends on whether the particles passing through the orifice are holes or electrons, e is the electric charge, V is the voltage drop across the contact, and p_F is the Fermi momentum. The excess current I is proportional to the current density times the area, or $ne\Delta v\pi a^2$, where n is the particle density. Calculating the resistance as V/I, we find $R = p_F/ne^2\pi a^2$. In the Drude theory of the electron gas the quantity p_F/ne^2 is equal to ρl, the resistivity times the effective scattering length. In the high K regime the resistance of an electrical contact has been estimated by Sharvin (*11*) as

$$R_S \cong \frac{\rho K}{4a} = \frac{K}{2} R_M = \frac{\rho l}{4a^2} \qquad (5)$$

(Note that in this form the resistance is quadratically dependent on the inverse of a.) In our experiments the Knudsen resistance is larger than the Maxwell resistance by the factor $K/2$, or about 100. Using the Sharvin formula, we find the (larger) estimate for a of about 40 Å.

Quantum effects. Thus, a is much larger than the de Broglie wavelength, so that direct quantum-mechanical Fresnel (edge-of-the-hole) interference phenomena (*12*, p. 161) would be weak, and would be visible only if a large voltage (about 600 mV) were impressed on the contact. However, if two or more of the "mountains" on the etched tip were of roughly equal height, but spaced "far" apart, then it would be possible to see the quantum-mechanical analog of Young (double-slit) interference (*12*, p. 114). If the two "mountains" were spaced 600 Å apart, then by the Heisenberg relation, an extra momentum p of about 10^{-22} gram-centimeter per second would be needed, or an excess energy of about 0.5 mV for free electrons, to see interference.

In a few of our formed contacts, we see exactly the same emission phenomena, as will be discussed below, but with a 60 percent (height) modulation at a "frequency" of about 0.75 mV. We interpret these as quantum interference effects due to multiple junctions. Such effects were also seen in the experiments of Yanson (*6*).

Interpolation Formula

Between the two regimes—low K (Maxwell) and high K (Sharvin)—one needs an interpolation formula which systematically treats the problem and yields these extreme limits. Wexler (13) has given such a formula, based on a variational solution to the nonlocal Boltzmann equation. He transformed the problem of electron flow through an orifice into the problem of an emitting disk. The transformation is similar to that used by Babinet in treating the analogous problem of the equivalence of screens and holes in geometric optics (12, p. 147). Wexler's interpolation formula is

$$R_{\text{int}} = \frac{\Gamma(K)}{2\sigma a} + \frac{4K}{3\pi\sigma a} \tag{6}$$

where $\Gamma(K)$ is a slowly varying function of K and takes the value 1 for $K = 0$ and $9\pi^2/128$ for large K. The first term is like the Maxwell resistance, the second like the Sharvin resistance. In our experiments we take the partial derivative of the contact resistance with respect to voltage V. We assume that the orifice radius is independent of voltage, and the scattering length is dependent on voltage (or energy, eV). The combination K/σ in the Sharvin-like term is independent of voltage since l/σ is independent of the scattering length. Thus only the first term enters the partial derivative

$$\frac{\partial R}{\partial V} = \frac{\Gamma(K)l}{2\sigma a}\frac{\partial}{\partial V}\left(\frac{1}{l(eV)}\right) \tag{7}$$

(Note that the structureless second term in Eq. 6 serves as a "series ballast resistor" to establish the voltage drop on the contact. The first term is the "active" one.) The total scattering length l is given by

$$\frac{1}{l} = \frac{1}{l_{\text{imp}}} + \frac{1}{l_{\text{ep}}} \tag{8}$$

where l_{imp} is the scattering length due to impurities, and where l_{ep}, the electron-phonon emission length, is given by

$$\frac{1}{l_{\text{ep}}} = \frac{1}{v_F\tau(eV)} = \frac{2\pi}{v_F\hbar}\int_0^\infty d\omega\alpha^2F(\omega) \times [2N_0(\omega) + 1 - f_0(\omega - eV) + f_0(\omega + eV)] \tag{9}$$

where v_F is the Fermi velocity, $\alpha^2F(\omega)$ the electron-phonon coupling, $1/\tau(eV)$ the energy-dependent emission rate, and $\hbar$ Planck's constant over 2π. Equation 9 is the condensed matter analog of Fermi's second golden rule. The "matrix element-squared" is α^2, the coupling constant, and $F(\omega)$ is the phonon density of states. The thermal Bose (N_0) and Fermi (f_0) factors enter because in condensed matter one needs to thermodynamically average over the system. At low temperatures, as in our experiments, the Bose factor vanishes and the Fermi factors are nearly step functions. Taking the voltage derivative, these become Dirac delta functions centered at $\omega = \pm eV$. In the low-temperature limit only the one at $\omega = eV$ contributes.

Table 1. Experimental parameters for the point contacts of Fig. 3. Symbols: R_0, resistance of the point contact at zero voltage; $k\theta_D$, Debye energy; a, radius of the contact, determined from R_0 $(4/3\pi)(\rho l/a^2)$; K, Knudsen number; λ_G, electron-phonon parameter recommended by Grimvall (15); λ_M, electron-phonon parameter determined by Hoyt and Mota (16); and λ_P, electron-phonon parameter determined by the point contact technique.

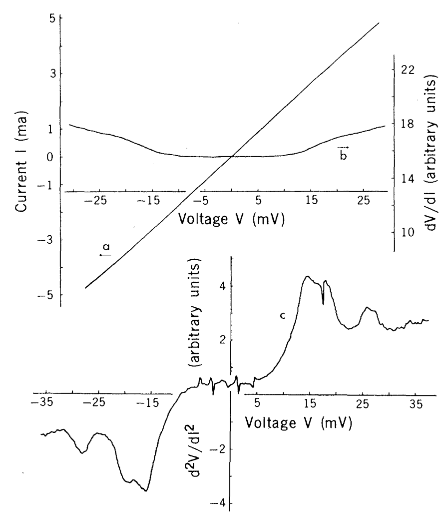
Material	R_0 (ohm)	$k\theta_D$ (meV)	a (10^{-8} cm)	K	λ_G	λ_M	λ_P
Cu-Cu	5.7	29.2	72.7	96	0.14 ± 0.03	0.16	0.14
Ag-Ag	16.3	19.1	54.9	140	0.10 ± 0.04	0.16	0.15
Au-Au	32.0	14.0	37.1	162	0.14 ± 0.05	0.21	0.16

Experimental Results

The data for copper plotted in Fig. 2, curve c, show both a slight asymmetry (about $V = 0$) and a slight positive shift in the signal. We attribute the asymmetry to a d-c voltage bias due to the thermal voltage over the coaxial leads to the point contact at 1.2°K. This small voltage shift is about 0.5 mV. We interpret the slight positive shift as due to first harmonic leakage into the second harmonic channel as well as tunneling leakage in the contact itself [which is even (8) in the second derivative]. The continuous data were digitalized [$\equiv D(\omega)$] on a grid of 0.5 mV. A new energy scale, shifted by Δ, was defined, and the symmetric and antisymmetric parts in the shifted energy scale were found $\{D_A(\omega) = ½[D(\omega - \Delta) - D(-\omega - \Delta)]\}$. The integral A_s of the antisymmetric part was defined as

$$A_s = \int_0^{\text{top}} D_A(\omega)d\omega \tag{10}$$

The shift Δ was varied in steps of 0.1 mV until A_s was maximized. This Δ was the same for all noble-metal point contacts. The resulting D_A's are shown in Fig. 3, together with literature values for the phonon density of states, obtained by fitting Born-von Karman models to inelastic neutron scattering data (14). The close coincidence of the peaks in the second derivative and the transverse and longitudinal peaks of the phonon structure suggest that bulk phonons play a



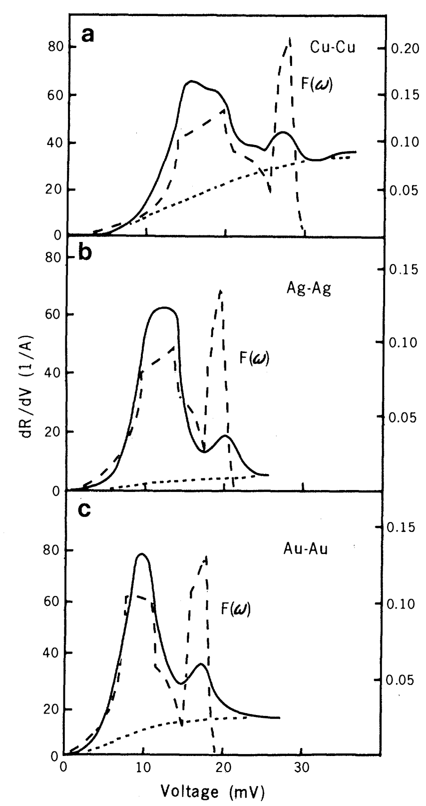


Fig. 2 (left). Photograph of the recorder sheet with (curve a) current I, (curve b) first derivative dV/dI, and (curve c) second derivative d^2V/dI^2 plotted against applied voltage. Fig. 3 (right). The measured voltage derivative of the resistance of point contacts as function of applied voltage for (a) Cu, (b) Ag, and (c) Au is plotted as solid lines (left-hand scale). The right-hand scale is the derived value for α^2F. The background functions are shown as short-dashed curves and the phonon density of states as long-dashed curves.

dominant role in the phenomena of point contacts. As the resistance at zero bias R_0 is varied, the ratio γ of the height of the first peak to that of the flat portion above the second peak varies inversely with $R_0^{1/2}$. Thus dR/dV contains two effects: α^2F and the effect leading to the relatively flat dR/dV above the phonon peaks. By making point contacts of extremely high R_0 (> 100 ohms), Yanson (*6*) has shown that the flat portion can be made small relative to the phonon effect. Because of its relative smallness at high R_0, we have called the flat portion the background (BG) effect. Yanson only used data with high R_0 to find α^2F's; we wished a more global treatment. After some experimentation we found that the functional shape

$$\mathrm{BG}(eV) = B \tanh^2(3\ eV/2k\theta_D) \qquad (11)$$

where k is Boltzmann's constant given in appropriate units and θ_D is the Debye temperature, worked well; B is a constant fit at 30 mV. This functional form yields the short-dashed curves in Fig. 3. The difference between dR/dV and the background is the derived absolute α^2F from these experiments. The right-hand scales of Fig. 3 are found from the values listed in Table 1 and the derived relation between dR/dV and α^2F. Apart from Yanson's values, we know of no other experimentally determined α^2F's for noble metals in the literature. To check our results we integrated to find λ as

$$\lambda = 2 \int_0^\infty \alpha^2F/\omega\ d\omega \qquad (12)$$

These values are listed in Table 1 as λ_P and are in close (± 15 percent) agreement with the literature values λ_G (*15*) and λ_M (*16*). Using the fitted background function, we could derive similar values

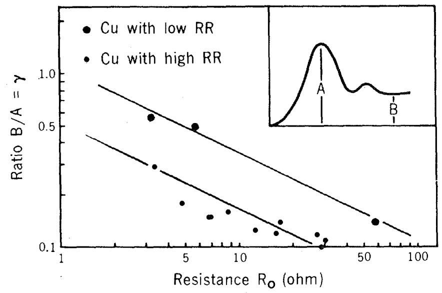


Fig. 4. The ratio B/A (the background constant divided by peak height) as a function of the resistance R_0 for two different values of the residual resistivity ratio.

of λ from dR/dV's for which R_0 ranged from 2 to 30 ohms.

On the basis of the coincidence of the structure of our curves in Fig. 3 with the structure of the phonon curves obtained from neutron scattering data, and the good agreement of our λ's with those from the literature, we conclude that the voltage derivative of the resistance of tiny point contacts (with background subtracted) provides a simple and convenient tool for measuring α^2F in normal metals.

Background Effect

We now examine the background effect in greater detail. In Fig. 4 we have plotted the logarithm of γ, the ratio of the background constant B to the peak height A of the derived α^2F, against the logarithm of R_0 for point contacts formed from copper having two different values of the residual resistivity ratio (RR). The large dots in Fig. 4 are for material whose RR was 71. The small dots correspond to samples formed from spears of RR 71 and anvils of RR more than 4000, with a geometric mean of 500. Plotted in this way, the data show that the background effect is related to the impurity concentration. The constant B varies as $\mathrm{RR}^{-0.5}$. With this separation by RR, the data in Fig. 4 are well fit by two lines of slope $-\frac{1}{2}$; that is, γ varies as $R_0^{-1/2}$.

We believe that the high current densities within the tiny contacts are beneficial in that electromigration effects will be swift: any impurity or defect will be rapidly swept away from the contact. In this sense, the contacts are "self-cleaning."

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