Negative differential resistance at atomic contacts

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Negative differential resistance is found for a pair of electrodes each consisting of a single Al atom coupled to a flat metal surface, where the Al atom is separated from the surface by a ''spacer'' atom (Br). This behavior is seen when the Al atoms are at a short distance from one another, and disappears when the distance is decreased to the point at which the Al atoms are in contact. [S0163-1829(97)09016-4]

Negative differential resistance in tunnel diodes was described long ago by Esaki,[1–3] and more recently, in the context of tunneling, has been commonly observed in quantum-well structures.[4] It was found in a calculation by Lang[5] to occur on an atomic scale in scanning tunneling microscopy, and was observed experimentally on this scale by Lyo and Avouris[6] and by Bedrossian, Chen, Mortensen, and Golovchenko.[7] We will see in the present paper how negative differential resistance develops at a pair of atom-sized contacts that has the appropriate density-of-states structure as the contacts are moved slightly apart.

The discussion on negative differential resistance at the atomic scale by Avouris and co-workers[6] points out that (just as in other tunneling contexts) whenever the two electrodes have relatively narrow density-of-states features of the appropriate energy (which sweep past each other as the bias is changed), it is possible for this effect to be present. Such narrow features can in general be obtained in the vicinity of the Fermi level by having a metal atom that is weakly coupled to the remainder of the electrode.[6,8] This can be accomplished most simply when the electrode is metallic by having adsorbed on it a ''spacer'' atom (or atoms), i.e., one with no states in the vicinity of the Fermi level, on top of which is a metal atom. Since metal-atom valence states will in general be at the Fermi level, the fact that the ''spacer'' atom has no states in this energy region means that the metal-atom valence resonance will be narrow.

To perform a calculation for a specific case, we take Al as the metal atom and a single halogen (Br) as the spacer. The configuration we consider is that shown in Fig. 1. A Br atom is adsorbed on each of the two flat semi-infinite metal surfaces at its estimated equilibrium distance[9] and the Al-Br bond length is taken equal to the sum of the covalent radii.[10]

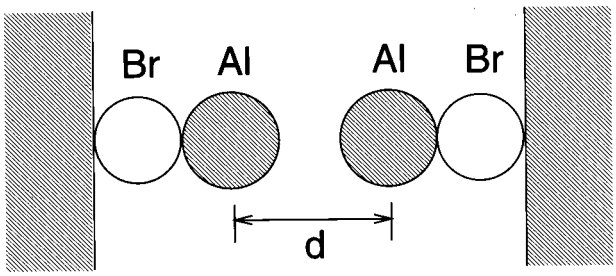

FIG. 1. Schematic diagram of the two-electrode system discussed in the text.

We will consider two values for d, the distance between the centers of the Al atoms: one equal to twice the covalent radius of Al, thus corresponding to contact (5.4 bohrs), and another several bohrs larger (8.6 bohrs). (1 bohr=0.529 Å.) All calculations are done fully self-consistently, with electron-electron interactions included, using the density-functional treatment described in Ref. 11. The electrodes are represented as in Ref. 11 using the uniform-background (jellium) model;[12] the cores of the atoms are represented using a pseudopotential.[13,14]

For purposes of discussion, we first show in Fig. 2 the density of eigenstates associated with the presence of a single Al-Br pair on one electrode.[15] We can think of changing the bias in the full system shown in Fig. 1 as sweeping two such densities of states (one associated with each electrode) past each other. Since there is a sharp Al-derived peak very close to the Fermi level (corresponding to $3p$ orbitals), we expect as discussed above the possibility of negative differential resistance behavior.

The current I at a given bias for the system pictured in Fig. 1 is obtained as discussed in Ref. 11, with I defined as the additional current that flows due to the presence of the atoms. (Note that the current per unit area in the absence of

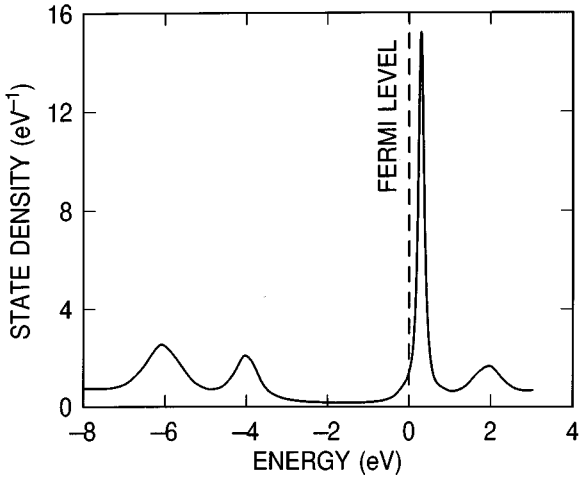

FIG. 2. Density of eigenstates associated with the presence of a single Al-Br dimer on one electrode (see Ref. 15). The upper three peaks correspond to Al orbitals, the lowest to Br orbitals. The Al peaks at -4, $+0.3$, and $+2$ eV correspond, respectively, to $3s$, $3p_{xy}$, and $3p_z$ states.

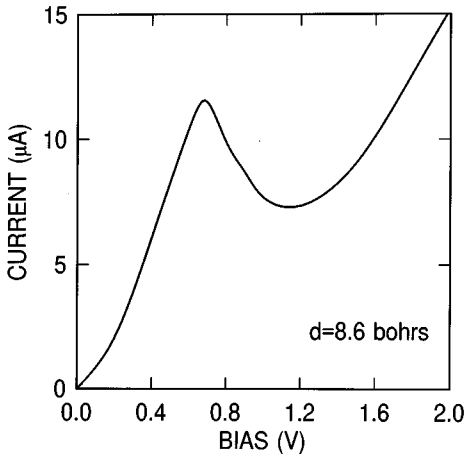

FIG. 3. Current-voltage curve for $d=8.6$ bohrs.

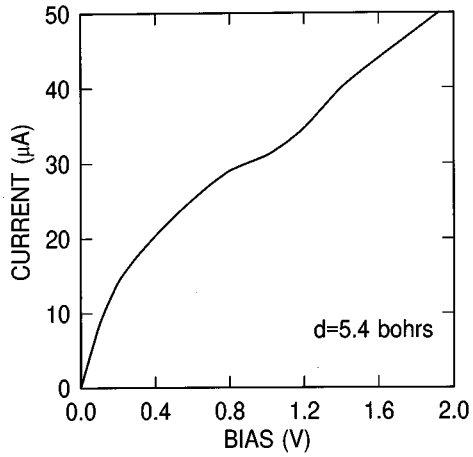

FIG. 4. Current-voltage curve for $d=5.4$ bohrs (corresponding to contact between the two Al atoms).

the atoms is negligible because of the relatively large distance between the electrodes.) This current is shown in Fig. 3 for the larger of the two spacings d considered; the plot does indeed show negative differential resistance behavior. Increasing the spacing d yet further should yield similar curves, but with a reduced current (closer to the experimentally accessible range).[16,17]

We now consider the case where d is reduced to twice the covalent radius of Al (i.e., contact). It is no longer possible to speak of sweeping past each other two narrow density-of-states structures, one of which is clearly associated with each electrode, and we thus expect the negative differential resistance effect to disappear. This is, as seen in Fig. 4, just what happens (there is only a slight concavity near 1.2 V, a small remnant of the minimum seen at this voltage in Fig. 3).

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[1] L. Esaki, Phys. Rev. **109**, 603 (1958).

[2] The importance of this effect in electronics is discussed, for example, by S. M. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981), Chap. 9.

[3] The general phenomenon of negative differential resistance was known long before the tunnel diode. See, e.g., H. T. Simon, Phys. Z. **6**, 297 (1905); E. W. Herold, Proc. IRE **23**, 1201 (1935); R. Landauer, Comments Solid State Phys. **4**, 139 (1972).

[4] For example, H. Kurata and H. Sakaki, Surf. Sci. **305**, 369 (1994); T. J. Shewchuk, P. C. Chapin, P. D. Coleman, W. Kopp, R. Fischer, and H. Morkoç, Appl. Phys. Lett. **46**, 508 (1985); T. C. L. G. Sollner, W. D. Goodhue, P. E. Tannenwald, C. D. Parker, and D. D. Peck, *ibid*. **43**, 588 (1983); R. Tsu and L. Esaki, *ibid*. **22**, 562 (1973).

[5] N. D. Lang, Phys. Rev. B **34**, 5947 (1986).

[6] I.-W. Lyo and Ph. Avouris, Science **245**, 1369 (1989); Ph. Avouris, I.-W. Lyo, F. Bozso, and E. Kaxiras, J. Vac. Sci. Technol. A **8**, 3405 (1990).

[7] P. Bedrossian, D. M. Chen, K. Mortensen, and J. A. Golovchenko, Nature **342**, 258 (1989).

[8] Cf. M. L. Yu, N. D. Lang, B. W. Hussey, T. H. P. Chang, and W. A. Mackie, Phys. Rev. Lett. **77**, 1636 (1996); C. B. Duke and J. Fauchier, Surf. Sci. **32**, 175 (1972).

[9] This is taken to be 1.8 bohrs, measured between the center of the Br atom and the positive-background edge of the uniform-background (jellium) model representing each electrode. It is obtained by using the layer spacing for Br adsorbed on Cu(111) determined by M. F. Kadodwala, A. A. Davis, G. Scragg, B. C. C. Cowie, M. Kerkar, D. P. Woodruff, and R. G. Jones, Surf. Sci. **324**, 122 (1995), together with the fact that the positive-background edge of a jellium is half an interlayer spacing outside of the center of the outermost layer of the metal it represents (neglecting contractions or expansions of the layer spacing at the surface).

[10] This is 4.9 bohrs; the covalent radii used are those given by P. R. Watson, M. A. Van Hove, and K. Hermann, *Atlas of Surface Structures: Volume IA* (American Institute of Physics, Woodbury, NY, 1994), Appendix D.

[11] N. D. Lang, Phys. Rev. B **52**, 5335 (1995). The success of the calculational procedure outlined in this paper is seen in the comparison between theory and experiment given by A. Yazdani, D. M. Eigler, and N. D. Lang, Science **272**, 1921 (1996).

[12] In the calculations, we will take the electrodes to have $r_s=2$ bohrs, typical of a high-electron-density metal such as Al. This parameter is defined by $(4/3)\pi r_s^3 \equiv n^{-1}$, where n is the mean interior electron number density in the electrodes.

[13] D. R. Hamann, M. Schlüter, and C. Chiang, Phys. Rev. Lett. **43**, 1494 (1979); G. B. Bachelet, D. R. Hamann, and M. Schlüter, Phys. Rev. B **26**, 4199 (1982).

[14] The number of plane waves is specified by $N=5$ for the two

transverse directions and by $N=9$ for the surface-normal direction (see Ref. 11) and the calculation is done in a rectangular box of width 12 bohrs in the transverse directions and 26 bohrs plus the excess of d over its value at contact, along the surface normal.

[15] The eigenstate density shown in Fig. 2 is the zero-bias density of eigenstates for the system consisting of the two electrodes with an Al-Br dimer attached to just one of them, minus the state density for the electrodes without the atoms. The eigenstates referred to here are those of the single-particle equations of the density-functional formalism. The electrode spacing is taken to be 22 bohrs (measured between the positive-background edges), which is the larger of the two distances used in the calculations with Al-Br dimers present on both electrodes. Since in this computation, the surface of the electrode without the dimer will be quite far from the dimer that is present on the other electrode, the density of states shown is essentially that which would be obtained if the bare electrode were absent entirely.

[16] It is difficult for computational reasons to do calculations at these very large distances, however. (Of course a tunneling-Hamiltonian formalism could be used.)

[17] With respect to the question of heat dissipation at larger currents, we note that this dissipation should occur primarily in the electrodes, and not at the atomic contact. [See S. Datta, *Electronic Transport in Mesoscopic Systems* (Cambridge University Press, Cambridge, 1995), pp. 69–72.] A piece of evidence that this is indeed the case is provided by a recent experiment in which a 2-μA current was passed through a single Mn adatom on a Cu(100) surface by a contacting scanning-tunneling-microscope tip, with the current remaining stable for several minutes [D. M. Eigler (private communication)].