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#### BARRIER PROFILES IN CLEAN AND DOPED TUNNEL JUNCTIONS

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Résumé.- Nous avons fait les calculs en utilisant la méthode de WKB et nous les avons fait accorder avec la conductance mesurée des jonctions tunnels (Al-I-Pb), les unes propres et les autres avec les. impuretés. Les barrières déduites pour les jonctions propres sont asymétriques ( $\phi_1 \approx 1,5 \text{ eV}$ ,  $\phi_2 \approx 4,5 \text{ eV}$ ). Nous expliquons ces résultats par une différence de concentration des espèces hydroxyles négatives à travers la barrière. Une barrière haute ( $\approx 10 \text{ eV}$ ) et mince ( $\approx 2,5 \text{ Å}$ ) décrit la couche organique des jonctions avec les impuretés.

Abstract;- We have measured the conductances of clean and doped Al-I-Pb tunnel junctions and fitted them by a WKB calculation in which the barrier parameters were varied. In undoped junctions the barrier so deduced is highly asymmetric ( $\phi_1 \approx 1.5 \text{ eV}$ ,  $\phi_2 \approx 4.5 \text{ eV}$ ). We explain the results by a concentration gradient of fractionally charged negative hydroxyl species in the barrier. A thin ( $\sim 2.5 \text{ Å}$ ) high ( $\sim 10 \text{ eV}$ ) barrier, associated with the organic layer, reproduces the measured conductances in doped junctions.

Fisher and Giaever /1/, in their pioneering paper, determined the current-voltage response of Al-I-Al sandwiches in the range 0 to ~ 2V. They found a roughly linear current increase at low (< 50 mV) bias, exponential increase at high (> 200 mV) bias and an exponential increase of resistance with insulator thickness. All these observations were shown to fit well to an analysis of tunneling due to Holm /2/. Comment was made on the inhomogeneity of the tunnel barrier, which was evident from a slight rectification present at all values of bias. The only surprise was a discrepancy between the Holm thickness required to explain the results and the values deduced by capacitance measurements. The latter were greater by a factor of three and the discrepancy was resolved by attributing an effective mass of 1/9 to the tunneling electrons.

Later, more extensive and reliable data enabled Rowell /3/ to identify a common feature of nearly all tunnel junctions : the conductance could be roughly approximated by a parabola with its axis offset from zero bias. Subsequently Brinkman, Dynes and Rowell /4/ (hereafter BDR) carried through a numerical evaluation of the conductance for a trapezoidal barrier shape. A problem which BDR encountered was the failure of their model to explain large ( $\sim$  250 mV) offsets of the conductance minimum. They suggested a more distorted potential was required to account for the behaviour and noted a correlation between the offsets and organic contamination of the junctions. We have measured the dynamic conductance of clean and deliverately doped tunnel junctions and attempted to determine the barrier parameters by fitting the results to the conductance calculated in the BDR fashion. The fits were found by trial and error variations of the parameters in the calculation.

The total current at bias V is

$$J(V) = \frac{4\pi me^2 V}{h^3} \int_{0}^{E_F} eV P(E_Z) dE_Z + \frac{4\pi me}{h^3} \int_{E_F}^{E_F} E_F eV$$

 $(E_{F}-E_{z})P(E_{z})dE_{z}$ 

For evaluation of the tunnel probability a trapezoidal barrier is chosen to represent the insulator in a clean junction. In doped samples an organic barrier of height  $\phi_a$  and thickness S is added.

Tunnel sandwiches of the type Al-I-Pb were fabricated by well-established techniques. A wide variety of organic species will chemisorb to the freshly grown oxide (Walmsley et al. /5/). By varying the exposure, any coverage up to saturation ( $\sim$  1 monolayer) is possible. The convention adopted in this work is that positive bias corresponds to the lead (Pb) electrode being positive.

An example of measured and fitted conductances is shown in figure 1. The fitting data for a number of samples is shown in Table I. Particular attention was paid to obtaining the correct absolute value of the conductance  $(\Omega_0^{-1})$  at zero bias and also at the bias limits.



 (a)
(b)
Fig. 1 : Trapezoidal and bilayer potential barriers representing clean and doped junctions.

#### Table I

DOPE	$\Omega_{o}$ (ohms)	φ,(eV)	φ,(eV)	d (Å)	φ <sub>3</sub> (eV)	S (Å)
	0					
NONE	23.6	1.57	4.70	11.60		
NONE	92.6	1.37	4.40	12.80		
NONE	567	1.47	4.70	13.39		
NONE	7250	2.28	4.07	14.43		
LAURIC ACID	62.2	0.40	4.90	7.56	10.70	2.90
ACETIC ACID	55.9	0.64	4.67	7.58	10.60	2.85
PHENOL	50.9	0.80	4.535	8.20	10.40	2.50
m-CRESOL	26.0	0.70	4.20	8.45	8.80	2.53
PHENOL (30%	) 202	1.50	4.70	11.00	10.50	1.00
PHENOL (90%	) 405	0.50	5.70	10.67	10.60	1.50

Even the clean samples show a strongly asymmetric barrier. In conjunction with evidence from the derivative of the conductance and capacitance measurements we attribute the asymmetry to a concentration gradient of fractionally charged negative hydroxyl species incorporated within the barrier during its growth.



Fig. 2 : Measured (solid line) and fitted (open circles) conductances of Al-I-Pb junction doped to saturation with lauric acid ( $\Omega_0$  = 62.2 ohms). Dashed line shows asymmetry of measured conductance.

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