ANOMALOUS CAPACITANCE OF THIN DIELECTRIC STRUCTURES

C. A. Mead
California Institute of Technology, Pasadena, California
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The classical analysis of a plane parallel capacitor gives a capacitance which becomes infinite as the spacing between the metal electrodes approaches zero. The fact that real capacitors show a marked deviation from this behavior has not generally been recognized.[1,2] This communication describes experiments in which the deviations have been measured with some care.

In order to determine the dependence of capacitance upon thickness it is necessary to have a reliable measure of the relative thickness of the device and also a method for producing very thin, uniform dielectric layers. Normal methods for measuring thickness are quite inconvenient for the thin layers used here. Consequently the voltage required to obtain a given tunneling current density[3,4] was used as a measure of the thickness. This procedure is considered to be a good approximation if the difference in metal-insulator work functions of the two metals under the conditions used is small, since the tunneling current density is only a function of the electric field at the interface, and not of the thickness of the dielectric. Investigation of the slight asymmetry of the volt-ampere characteristic of the structures used indicated that differences in the metal-insulator work functions were indeed small.

The present experiments were all carried out on a tantalum sheet which had been recrystalized in a dry argon atmosphere at 2700°C for 1/2 hour. The sheet was then composed of many crystals approximately 1 mm across, and was subsequently electropolished to a mirror finish. Tantalum oxide was formed on the surface anodically in dilute ammonium citrate solution. The samples were then placed in a vacuum system where they were baked at 300°C at residual pressure of 3×10^{-9} mm Hg while counter electrodes of gold were evaporated through a mask. The resulting structure was a large number of gold squares approximately 5 mils (0.005 in.) across on the tantalum oxide surface. Measurements of capacitance and tunnel voltage were made by standard techniques on a large number of squares on each sample. Only squares lying wholly on one crystal were used. The tunnel current density used was approximately 0.05 ma per square mil. Previous experience had indicated that a very small amount of contamination of the surface resulted in large variations of capacitance between squares on one sample. On the samples tested it is believed that surface contamination has essentially been eliminated since variations were in many cases less than 1 % and in all cases were well within the limits placed upon the measurement of the variation of area between squares. The results of this experiment are shown in Fig. 1. It is seen that instead of having zero intercept, the plot of inverse capacitance versus tunneling

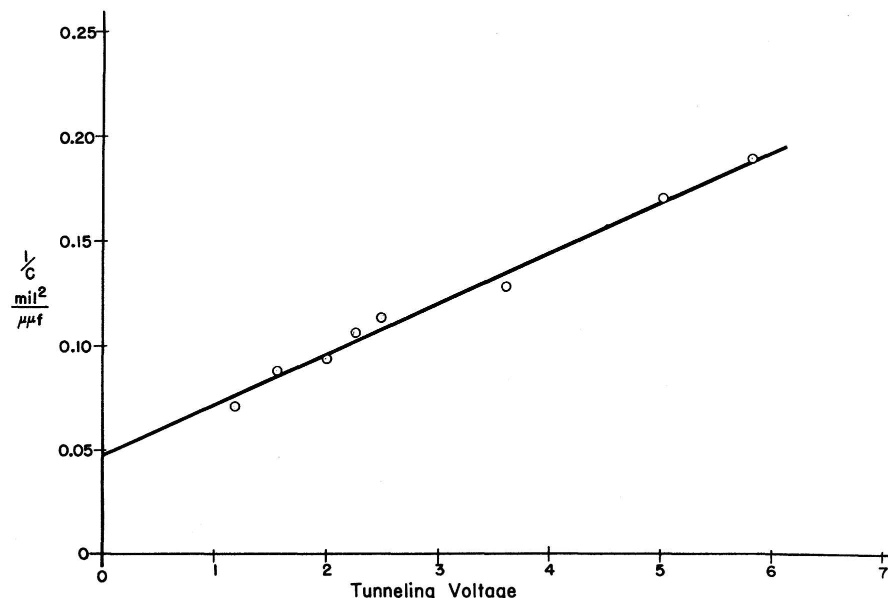

FIG. 1. Inverse capacitance as a function of tunnel voltage for $Ta-Ta_2O_5-Au$ structures.

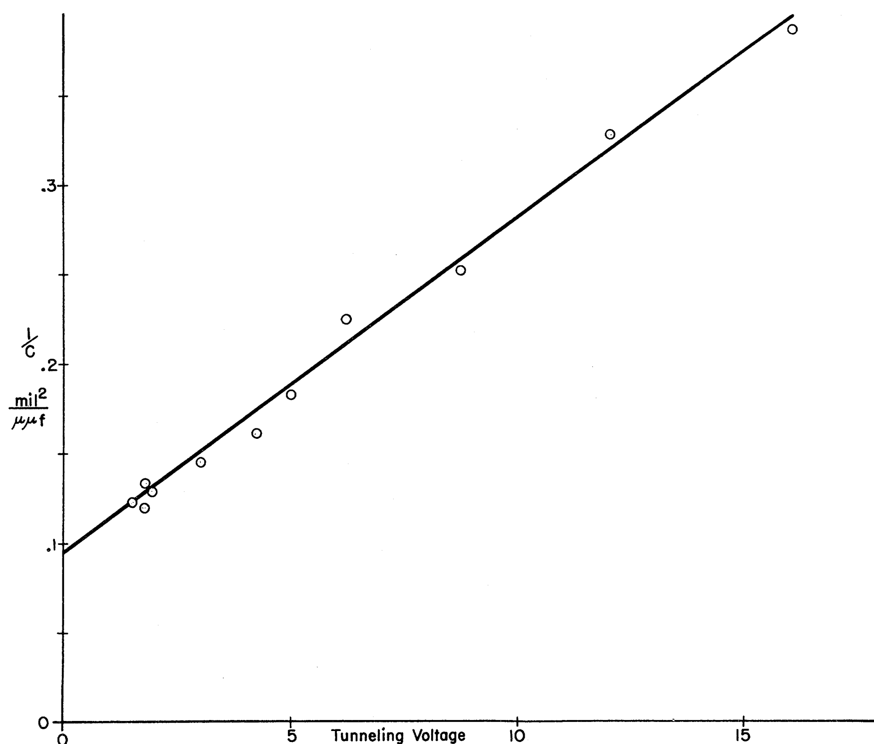

FIG. 2. Inverse capacitance as a function of tunnel voltage for Ta-Ta_2O_5-Bi structures.

voltage intercepts at approximately 0.05 square mil per $\mu\mu$f. This intercept corresponds to an ϵ_0 layer approximately 2.75 A thick. It was felt that perhaps variations in the capacitance with applied dc electric field could be observed. The experiment was performed but no detectable variation was observed up to electric fields where tunneling current made the measurements unreliable.

Theoretical work[5] indicates that the dielectric constant of an insulator should be preserved down to a few atomic layers. If this is true, we should expect the basic limitation of the capacitance to be electric field penetration into the metal. A crude classical calculation using a method similar to that of Mott and Jones[6] gives a penetration depth of the order of one A for typical metals, varying inversely with the square root of the density of states in the metal. Although no real confidence can be placed in such a calculation, if this is indeed the mechanism of the capacitance limitation, the $1/C$ intercept should vary with the density of states in the metal.

This dependence was investigated by another set of experiments using bismuth counter electrodes. Because of its low density of states, bismuth should have a larger value of penetration depth and hence a higher intercept on the plot of $1/C$ versus tunneling voltage. That this is indeed the case is shown in Fig. 2. Here the intercept corresponds to an ϵ_0 layer approximately 5.5 A thick.

At present it has not been possible to achieve truly symmetric structures and hence the series capacitance contribution for each interface is not known, but only the sum of those for two interfaces. A symmetric structure plus a knowledge of the true dielectric constant within the metal would be necessary for a full determination of the parameters involved.

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