FINAL TECHNICAL REPORT

ONR GRANT INFORMATION

Grant Title: **High Resolution Studies of Thin Film Interfaces**

Performing Organization:	Cornell University
Principal Investigator:	R. A. Buhrman
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REPORT SUMMARY

This project was concerned with high-resolution studies and applications of ballistic electron transport phenomena, and with microscopic investigations of the interfacial properties of thin film electronic systems. The principal tool in this investigation was ballistic electron emission microscopy (BEEM), and related scanning tunnel microscopy (STM) measurements. The focus of the project was on the study of the possible effect of energetic electrons (~ 2 - 5 eV), tunnel-injected into thin film electronic materials from STM tips, in locally stimulating the formation of atomic vacancies and defects, and in the subsequent promotion of interdiffusion of atoms at electronic interfaces, with one objective being the development of new ways to manipulate the transport and other properties of materials at nanoscale dimensions. Momentum dependent BEEM measurements in epitaxial systems was also employed in the study of interfacial transport and scattering in metallic multilayer systems of potential technological importance.

Description of the Scientific Research Goals of this Grant

The plan for this experimental research program was based on the utilization of scanning tunneling microscopy and ballistic electron emission microscopy to induce and examine changes at Schottky barrier and thin film interfaces that can be generated at the nanometer scale by energetic electron injection. We also sought to pursue BEEM studies of ballistic transport in thin film multilayer systems with emphasis on examining the consequences of scattering and of surface and bulk electronic structure on this transport.

The project had two main elements:

1: The extension of our previous study of the energetic-electron-stimulated formation of adatom/vacancy pairs at Au/Si interfaces to different, and more stable Schottky barrier systems. The objective was to utilize the ability to locally create non-equilibrium vacancy concentrations by hot electron injection in the study of inelastic scattering processes from atomic defects, and to stimulate and examine the consequences on electronic transport of the intermixing of thin film interfaces.

2: The utilization of the momentum selective aspect of ballistic electron transport of Schottky barrier systems as a new means of studying important aspects of interfacial transport in thin film multilayers. The project emphasis was on the application of this technique to the investigation of band structure effects on ballistic transport in metallic thin film systems, and on the characterization of the interfacial transport properties of metallic interfaces of the type that are the building blocks of technologically important thin film multilayer structures, such as those that exhibit the giant magnetoresistance effect.

Background Information

This project was originally intended to extend from 11/1/95 through 10/30/97. Due to staff problems, and more difficult than anticipated technical challenges, progress on the project was initially not as rapid and substantial as expected. Therefore, by mutual agreement between the Principal Investigator and the Scientific Program Officer, funding for this project was terminated at the end of the first year of funding, 10/30/96. However, the project was able to move forward with support from other sources, principally in the form of graduate fellowship support, and substantial progress was made, which built on the groundwork that had been laid earlier. This report discusses the results of the initial instrument development effort and the highlights of the subsequent research that have resulted from that work.

Significant Results Achieved Under the Contract

The first step in this project was the completion and commissioning of an advanced, high performance UHV scanning tunneling microscope/ballistic electron emission microscope (STM/BEEM) system whose construction had been initiated during the previous ONR grant. While this effort was more time-consuming than expected, the resulting instrument, which is depicted in Fig. 1, has proven to be excellent. It has a higher measurement bandwidth than any BEEM instrument previously reported. It is also very stable and yields excellent UHV BEEM images, with much lower drift than indicated by typical BEEM images shown in the literature that have been taken with other instruments. This BEEM system has also proven to be quite versatile, permitting straightforward and rapid tip exchanges and sample interchanges in UHV, which has greatly facilitated our experimental program.

The first application of this new system was to continue the study of hot-electron effects on the Au-Si Schottky barrier systems. In previous efforts we had found that high energy, >2 eV, ballistic electrons injected from the STM tip could stimulate the formation of Au vacancy-adatom pairs at a Au-Si interface which had been passivated by depositing a monolayer or two of C onto the Si surface prior to the Au deposition. If a sufficient number of such adatoms were generated the passivated layer could be thinned locally, leading to an enhanced Schottky barrier transmissivity. On the other hand if the vacancy formation rate was high enough, the increased elastic scattering arising from these vacancies which would diffuse into the bulk of the Au would strongly attenuate the ballistic electron current. Our previous work had investigated this phenomena with measurements made in air, our first goal here was to establish whether a more controlled, UHV environment would alter the results. Qualitatively we found the results in UHV to be the same as observed earlier. Quantitatively, however, the much higher purity Au films that were formed in the UHV system did alter the details of the vacancy diffusion rates in the Au, which resulted in a faster annealing away of the Au vacancy clusters, making the hot electron "writing" even more transitory than that observed in air.

Our next planned step was to extend these hot electron studies to other Schottky barrier systems, and also to the examination of possible hot-electron induced mixing of metallic bilayers. In the first instance the objective was to see if local hot-electron modifications of Schottky barriers could be made permanent by the use of metal overlayer systems with much slower diffusion rates at room temperature. The approach was to heat the sample to an appropriate temperature, so that the thermally assisted vacancy formation could be stimulated during hot electron stressing, and then cool the sample down before the vacancies could anneal away. In the second instance the objective was to see if it were possible to stimulate local intermixing of thin metallic bilayers is a manner that would substantially affect the BEEM current to an underlying Schottky barrier interface. Unfortunately, in our efforts to date we have not yet been successful in finding a workable operating temperature window that would facilitate these results. We have had some tentative indication that hot electrons can indeed stimulate local intermixing, a result which we hope to confirm and further study in subsequent work. However, the most important development resulting from this investigation was the realization in the course of these experiments that the issue of ballistic transport in the metallic overlayers was much more complicated than had been assumed in the standard BEEM models. This realization resulted from some striking BEEM observations that are discussed below, and led us to pursue the implications of this fact as a main focus of our experimental effort.

In the original development of BEEM it was assumed that the metallic overlayer could be effectively modeled as a free electron system, ignoring the band structure in the metal. This was realized to be too simplistic in the case of the silicides, but for "free electron" metals such as Au the assumption seemed reasonable. Yet it is well know that even for Au, Ag and Cu, the band structure has complexities that can affect electron motion. In particular the Fermi surface in the (111) crystalline direction for these fcc metals is such that conduction electron waves cannot propagate in that direction. This results, for example, in the formation of surface-confined electron states on the (111) face of Cu or Au crystals. This was the basis of the recent and very beautiful "quantum corral" studies of Eigler and co-workers of standing wave resonances arising from such surface states. Yet despite the non-free-electron band structure of the noble metals, and the well-known existence of these surface states, and despite the fact that Au or Cu tends to grown preferentially on Si in the (111) normal orientation the implications of this band structure on BEEM measurements has been largely ignored apart from recent theoretical discussions by Garcia-Vidal and co-workers.

Since much attention has been paid to the issue of momentum conservation in electron transport across the Au-Si(111) and Au-Si(100) interfaces, it might appear surprising that a possible effect of the Au band structure has not been more seriously considered. But this is simply because up until now there has been no clean experimental indication that the metal band structure mattered. We now have strong experimental evidence that the band structure does matter a great deal.

The BEEM samples from which we obtained this evidence were Au-Si and Cu-Au-Si samples where the Au or Cu-Au bilayers were deposited in UHV onto very clean and H passivated Si surfaces. X-ray diffraction revealed that the crystal orientation of the Au or Cu-Au bilayers was more than 99% (111) normal for both the cases of growth on Si(100) and Si(111). Perhaps due to their UHV growth conditions the ballistic electron mean free path that we found in these samples by measuring the BEEM current as a function of metal thickness was much longer than previously reported from, presumably, more defective samples. For example, as shown in Fig. 2, in the case of the pure Au samples attenuation lengths of 220Å and 230Å were measured for Au on Si(100) and Si(111) wafers respectively. This is in contrast to, for example, recent measurements by Bell of an attenuation length of 130Å for his Au-Si BEEM samples for both Si orientations.

In addition to showing the large attenuation lengths for our samples, Fig. 2 also reveals the initially surprising result that for a given Au thickness the BEEM current is found to be consistently higher for Au(111) on Si(111) than it is for Au(111) on Si(100). Since electrons entering into Si(111) must have substantial transverse momentum to be permitted to enter at the conduction band energy minimum if transverse momentum is conserved in the process, it was initially expected that Au-Si(111) would have a much lower BEEM transmissivity than Au-Si(100). However, as first pointed out by Garcia-Vidal et al., if the Au band structure forces the electrons to propagate at a angle away from the normal to the surface, this would not be the case. Indeed our own simple model calculations suggest that since the band structure should constrain the ballistic electrons in clean Au to approach the Au-Si interface with substantial transverse momentum, the Au-Si(111) interface should have in fact a higher transmissivity in the absence of interfacial scattering than does the Au-Si(100) interface. This is what we observe.

An even more striking demonstration of the impact of the metal band structure on ballistic transport is seen in the BEEM behavior of Cu-Au bilayers deposited on Si. These bilayers were first made to study hot electron induced mixing, but our experimental focus quickly changed once we saw the results such as are illustrated in Fig. 3. When Cu in deposited on a thin Au overlayer on Si, the Cu atoms grow very flat and clean crystallites as can be seen in the STM image. Generally the surfaces of these grains show clear atomic steps, and of course low angle grain boundaries are also present in large quantities. What is particularly notable in the BEEM image, as shown in Fig. 3 also, is that there is a generally enhance BEEM signal at all of these surface steps and grain boundaries. The essentially 100% correlation between the locally enhanced BEEM signal, which can be as much as 50% or more in magnitude, is demonstrated more clearly by Fig. 4. That figure shows the same STM image as in Fig. 3, but after it has been processed to be the brightest at regions where the surface topography is changing the most rapidly. The one to one correspondence with the bright regions of the BEEM image in Fig. 4 is clear.

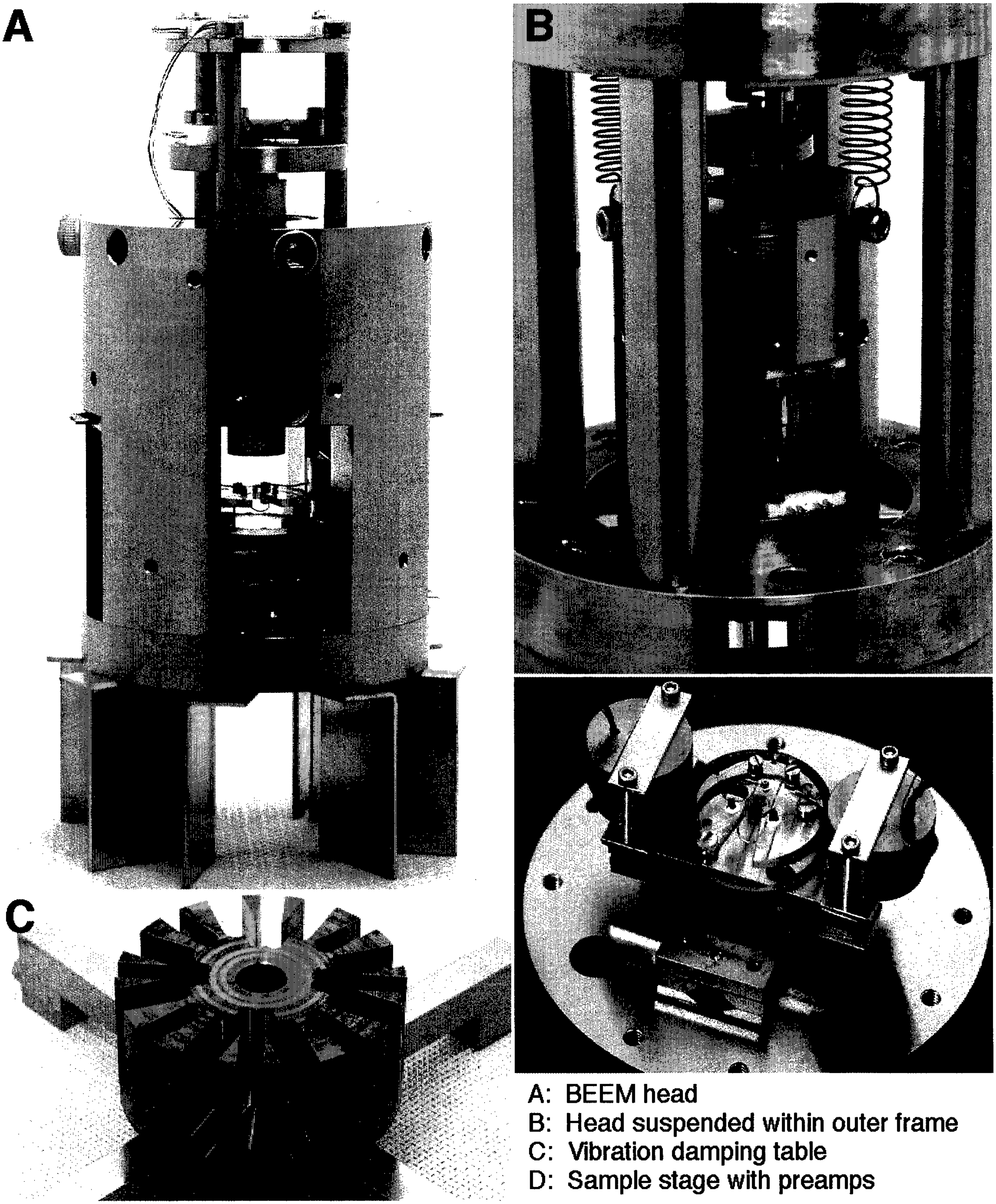
Our interpretation of Figs. 3 and 4 is that when the STM tip is positioned above the a flat (111) Cu surface the electrons are tunneling predominately to empty surface electron states which, due to the Cu band structure, are decoupled from the states that can propagate into the bulk. Only by an inelastic scattering process can electrons that are injected into the surface states make a transition to a bulk state. But such an inelastic scattering will reduce the electron energy and degrade the BEEM current. Thus the BEEM current is a measure of those electrons that directly tunnel into bulk states from the STM tips. At surface steps and grain boundaries the probability of tunneling directly to bulk states is much greater, due to the local topography, and thus the BEEM current is enhanced at these positions.

We find these to be very exciting results and are now actively pursuing additional experiments that will further study and elucidate this phenomena. One critical aspect that we are examining now is the role of the STM tip, e. g. its sharpness which will affect the angular momentum distribution of the tunneling electrons, in defining the portion of electrons that tunnel into empty surface states, which are distributed in a narrow cone of momentum about the normal to the substrate, and the portion that tunnel into empty bulk states. The BEEM measurements also appears to now give us a direct way to measure the coupling between the surface and bulk states. For example with respect to Eigler's quantum corral experiments, which were performed by arranging Fe atoms in a ring on a (111) Au or Cu surface to set up surface standing waves, there has been an unresolved issue whether the confining Fe atoms scattered the surface waves into other surface states or into bulk states. By measuring the BEEM current for such structures we should be able to measure directly how effective surface impurities are in scattering surface electrons into the bulk states. This is both a fundamental and technologically important issue since, for example, in giant magnetoresistance systems, surface and interfacial impurity atoms and defects could be very important in determining the electron transport properties of such systems. Indeed we see a number of exciting opportunities to employ BEEM of this type in the study of very important nanoscale transport issues, particularly in heterogeneous magnetic systems which are currently the focus of so much technological interest.

In summary, while this project did not initially make the rate of progress that was expected and thus came to an early termination, it did eventually lead to some very interesting and exciting results. We have successfully constructed an improved UHV BEEM system and have used it effectively to identify and study the effect of the metallic band structure on ballistic electron transport in Au-Si Schottky barrier systems. Most importantly we have also seen direct evidence of STM tunneling to surface states on Cu (111) affecting the BEEM transport to an underlying Schottky barrier. By extending these type of studies to, for example, Co-Cu multilayers we now hope to get new insights into the role of band structure and scattering in determining the critical details of ballistic transport in heterogeneous thin film systems.

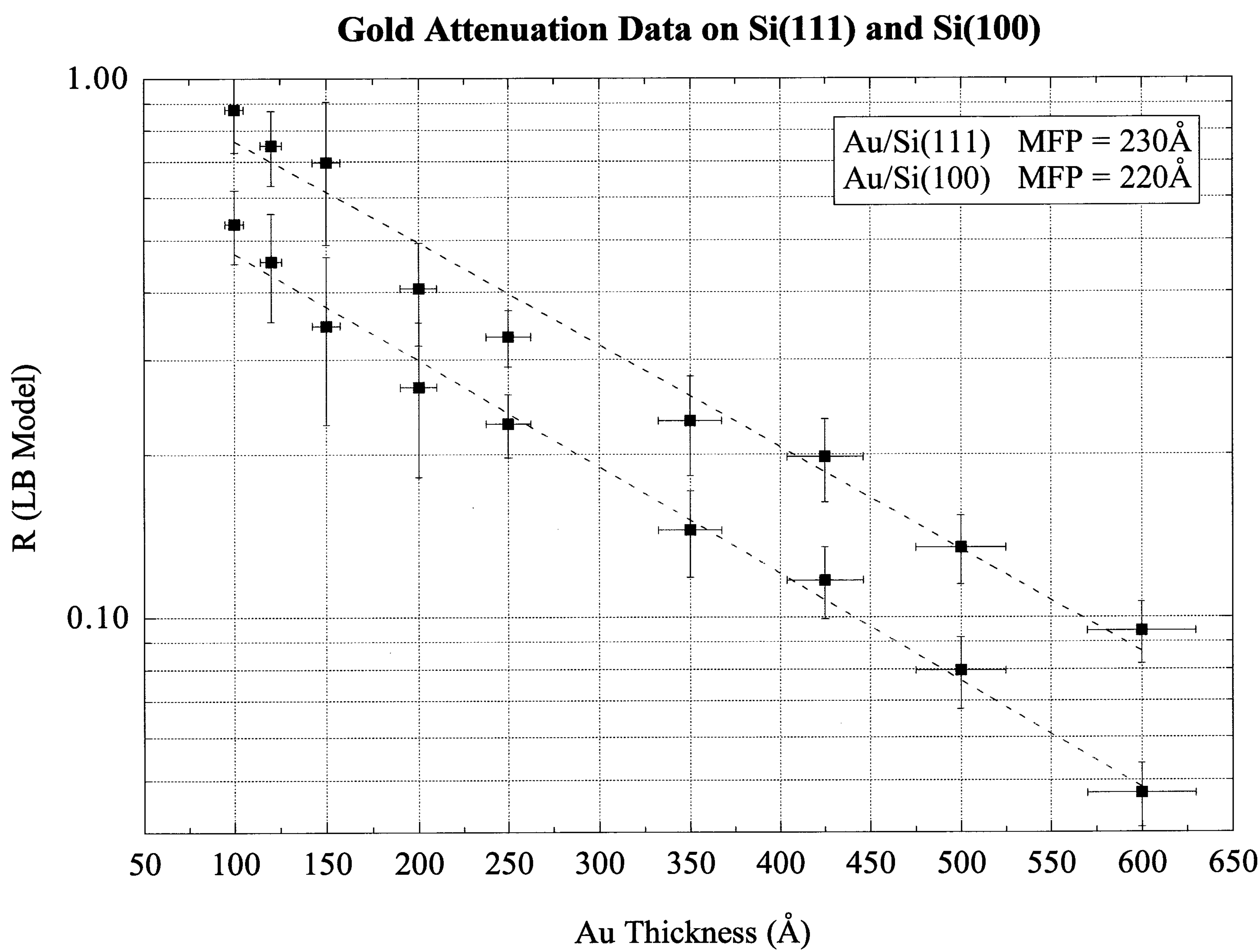
Papers Published in Refereed Journals Describing Work Supported in Whole or in Part by this Contract

None at this time, but several papers in preparation.



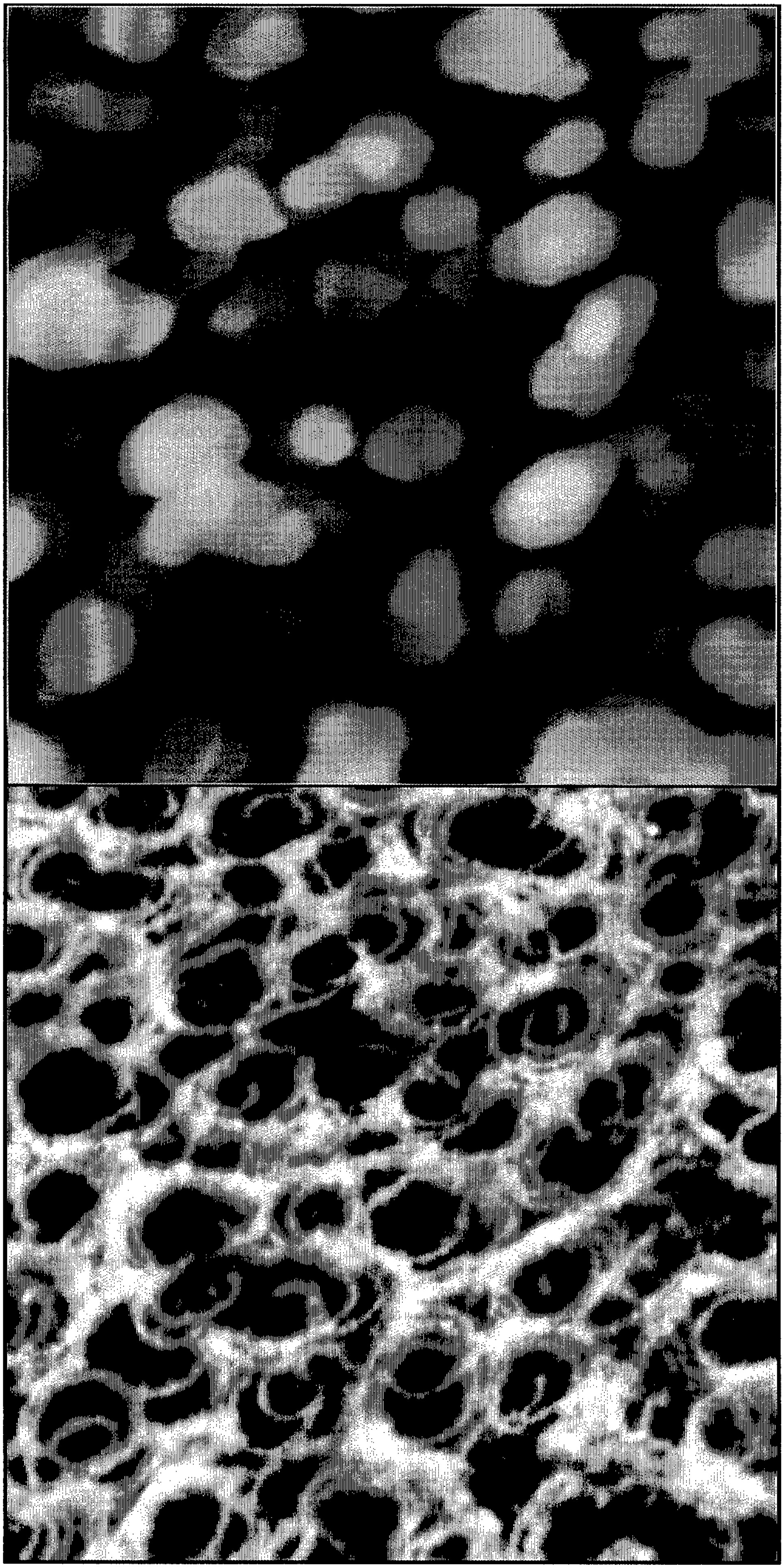
A: BEEM head
B: Head suspended within outer frame
C: Vibration damping table
D: Sample stage with preamps

FIGURE 1



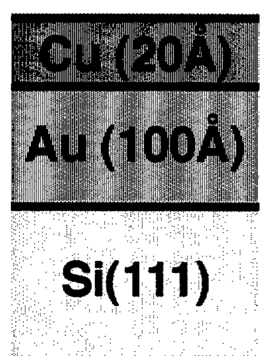
Gold Attenuation Data on Si(111) and Si(100)
R (LB Model)
1.00
0.10
Au/Si(111) MFP = 230Å
Au/Si(100) MFP = 220Å
50
100
150
200
250
300
350
400
450
500
550
600
650
Au Thickness (Å)

FIGURE 2



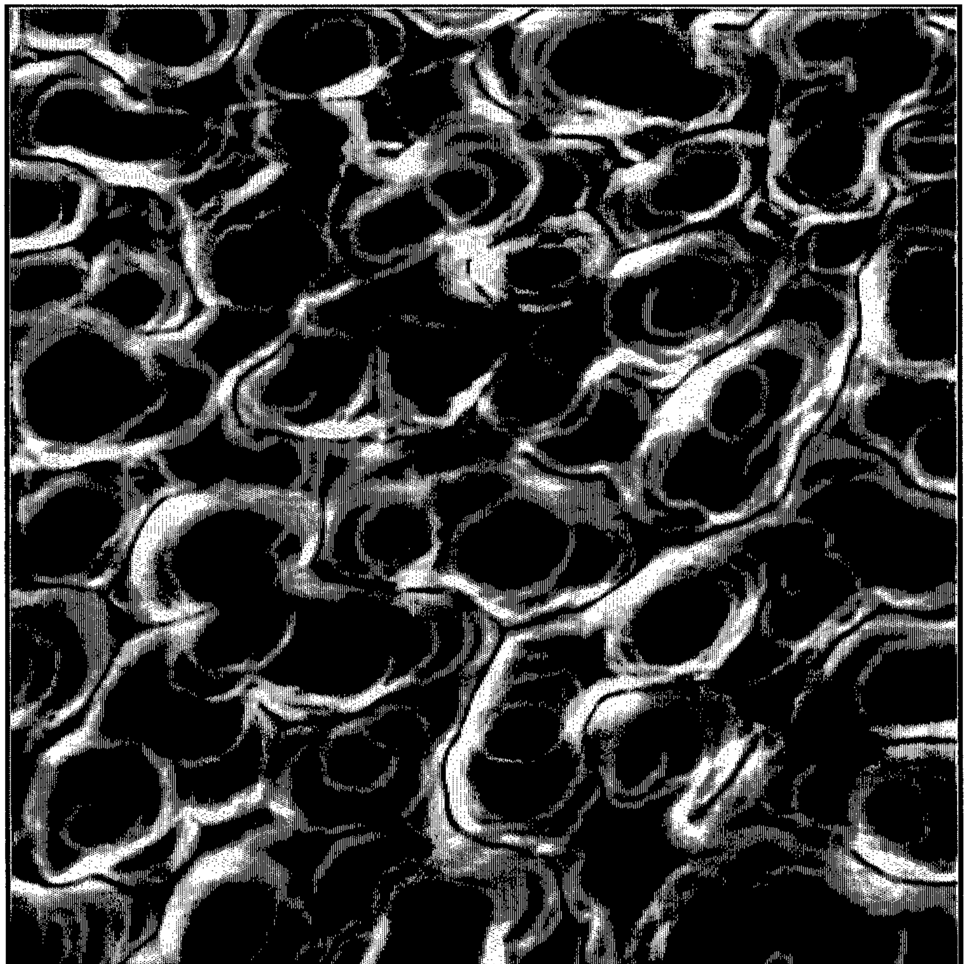
Surface Image
white: high
black: low
range: 16Å

BEEM Image
white: 10 pA current
black: 5 pA current

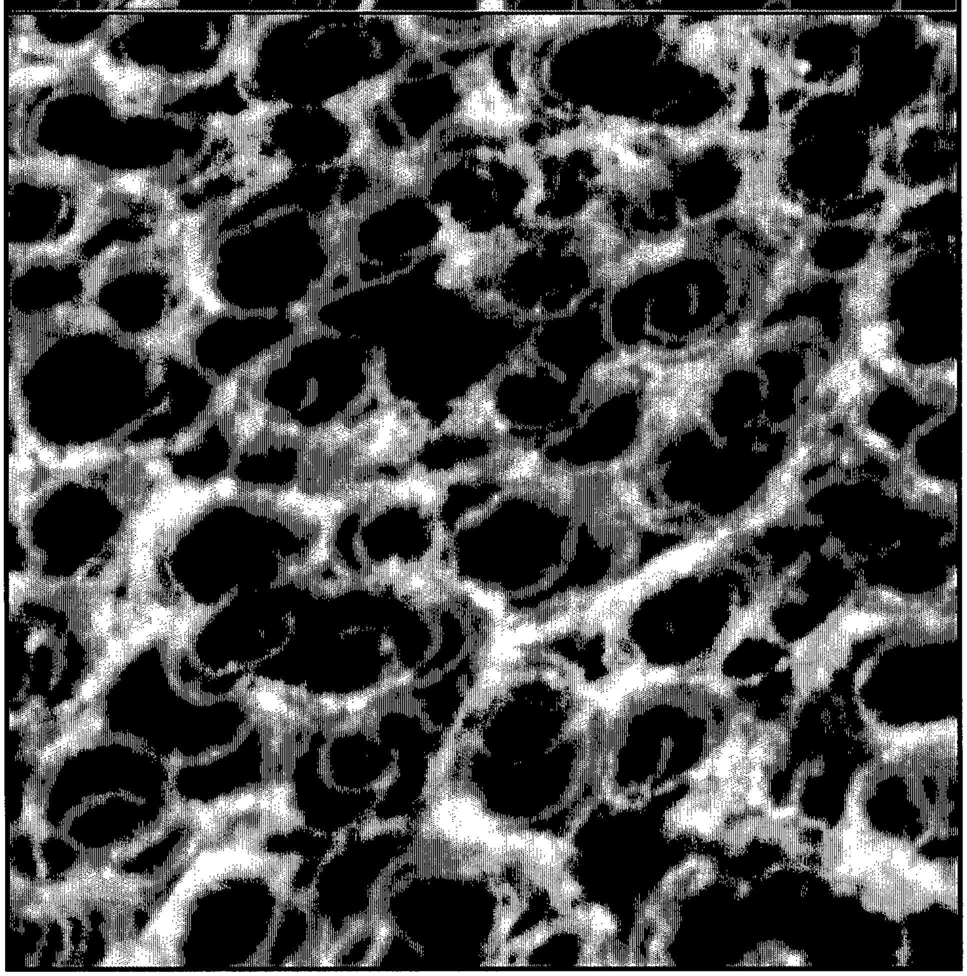


(100nm x 100nm)

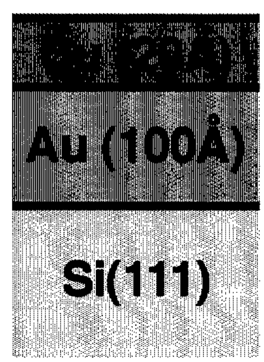
FIGURE 3



Surface Slope Image
white: steep slope
black: flat region



BEEM Image
white: 10 pA current
black: 5 pA current



(100nm x 100nm)

FIGURE 4

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13. ABSTRACT *(Maximum 200 words)*

This is the final technical report for a research project that has been concerned with high resolution studies of ballistic electron transport phenomena in thin film electronic systems, and with the study of hot electron effects on such systems. The principal tool in this investigation was ballistic electron emission microscopy (BEEM), and related scanning tunnel microscopy (STM) measurements. One focus of the project was on the study of the possible effect of energetic electrons, ~ 2 - 5 eV in energy, tunnel-injected into thin film electronic materials from STM tips, in locally stimulating the formation of atomic vacancies and defects, and in the subsequent promotion of interdiffusion of atoms at electronic interfaces. An objective was the development of new ways to manipulate the transport and other properties of materials at nanoscale dimensions. In addition, the investigation developed striking evidence of the critical role of surface and bulk electronic structure, even in comparably simple "free electron" metal thin films, in determining the direction and magnitude of ballistic electron transport in such materials.

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