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13. ABSTRACT (Maximum 200 words) This project is for purchase and installation of a high resolution analytical transmission electron microscope. The microscope has been delivered, installed in a specially prepared laboratory area, and thoroughly checked out. It met all performance tests and was formally accepted on September 1, 2003, two weeks after the final peripheral hardware was installed by the manufacturer and checked out successfully. The start date for the project was June 1, 2000. The microscope chosen for purchase after a rigorous evaluation process was a JEOL 2010F. It is a 200kV TEM/STEM microscope, capable of being fitted with electron energy loss and energy dispersive characteristic x-ray spectrometers, and special STEM detectors, to transform it from an ordinary transmission to an analytical scope. The basic microscope was delivered on September 28, 2001, from Japan, where it was manufactured. It was modified at the factory during construction so that two Seiko Turbomolecular vacuum pumps evacuate the column and viewing chamber. Ion pumps evacuate the field emission electron gun. Scroll pumps are used for all backing. Thus the vacuum system is completely dry with no oil diffusion or oil-sealed rotary mechanical pumps used. This was a new procedure adopted by the manufacturer, after much discussion with us and a colleague at AT&T Bell Labs, to minimize carbon contamination on the specimen during small probe STEM analytical operation. It was successful, and is now offered as a special extra cost option on currently ordered scopes.				
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September 18, 2003

Re: Final Report: Project F49620-00-1-0324, "High Resolution Chemical
Microscopy for Materials Science".
Project Director: R. W. Carpenter;
Co-investigators: Profs. W. Petuskey and D. J. Smith

This project is for purchase and installation of a high resolution analytical transmission electron microscope. The microscope has been delivered, installed in a specially prepared laboratory area, and thoroughly checked out. It met all performance tests and was formally accepted on September 1, 2003, two weeks after the final peripheral hardware was installed by the manufacturer and checked out successfully.

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Recently it became well known that STEM microscope performance is usually limited by the environment where they are installed. The major problems are electrical and mechanical disturbances that cause sub nanometer instabilities in the small probe position during scanning operation, and thermal transients that cause specimen drift, again seriously compromising spatial resolution in small probe analytical modes of operation. We addressed the mechanical vibration issues several years ago, by installing a special foundation in the room where the microscope is installed. It is separated from the main building foundation by a membrane, and proved successful in isolating the scope from mechanical vibrations. Other measures were required for isolation from electrical and thermal disturbances. After site stability measurements and discussions with colleagues who are facing the same problems at Cornell University, Bell Labs, Oak Ridge National Laboratory, and the University of Dresden, we decided

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on several courses of action to stabilize our site and pursued them to completion. They have been successful. They required an additional ~ \$ 100,000 expenditure by our University. The microscope has attained 0.14 nm resolution in STEM mode as a result. It is unlikely that this performance could have been reached without the environmental enhancements made to the site. According to the manufacturer, our site is the most stable of their installations, and they now use it as their best example of a successful site. The most unusual and costly site stabilization measure we used is a completely separate air conditioning system (from the building system) for the microscope room itself. The linear flow rate of cooled air in the room is less than one foot per second, and the room temperature is stable within 1 degree Centigrade with the room door shut.

The major peripheral analytical equipment on the microscope is a Gatan Enfina electron energy loss spectrometer, an EDAX Phoenix System energy dispersive characteristic x-ray spectrometer, several dark-field and bright-field STEM detectors from the JEOL, Gatan, and Fischione companies, and several CCD and television cameras from Gatan. Three external PCs and one internal computer are used to operate the microscope. The spectroscopy performance of the microscope has been excellent. The energy resolution of the Enfina spectrometer with the microscope in STEM mode (0.26 nm FWHM probe) is ≈ 0.9 eV with probe current of 50 Pico amperes, which has proven quite sufficient for spectroscopy with $\sim .5$ nm spatial resolution. The energy resolution under these analytical operating conditions is excellent, and actually better than expected for a Schottky field emission gun. Some examples of the performance are shown in the figures below.

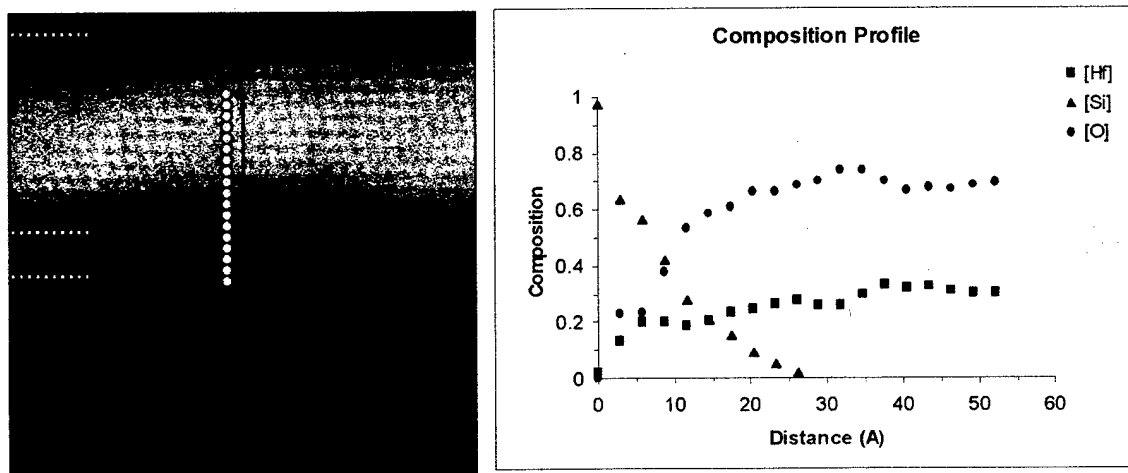


Figure 1. Left: Annular dark field (ADF) STEM image of two-nanolayer high dielectric constant stack made by atomic layer chemical vapor deposition. The bright top layer is mainly crystalline hafnium dioxide. The intermediate dark featureless layer about 1 nm thick (marked by the two horizontal dotted lines in the middle) is amorphous substoichiometric silicon oxide. The crystalline silicon substrate is visible at the bottom, viewed along a $\langle 011 \rangle$ direction. Two sets of Si $\langle 111 \rangle$ planes are visible edge-on in the substrate. Their spacing is 0.314 nm. The vertical line of dots shows the beam positions where energy loss spectra were acquired, to derive the composition plots shown at right. The arrow shows the direction of the probe trace during spectrum acquisition. Right: Plots of Hf, Si, and O composition across the multilayer stack shown. Zero on the horizontal distance scale corresponds to the Si substrate/Intermediate layer interface. Note the clear evidence of interdiffusion of Si and Hf across the intermediate layer/HfO_x interface.

ADF STEM images are created by collecting high-angle elastically scattered electrons; hence they are atomic number sensitive, and produce highest contrast in images of specimen regions that contain high atomic number elements, such as Hf. They are "chemically sensitive", but not spectroscopic. To determine chemical composition, a spectroscopy such as electron energy loss, used to derive the curves shown on the right of Figure 1, must be used. A major issue then becomes spectroscopic spatial resolution, which is mainly determined by the incident electron probe size at the specimen. This must be quite small for the research shown above, and it must contain sufficient current to acquire the individual spectra quickly (to avoid drift), as shown in Figure 2. The open dots define the experimentally measured distribution. The solid line

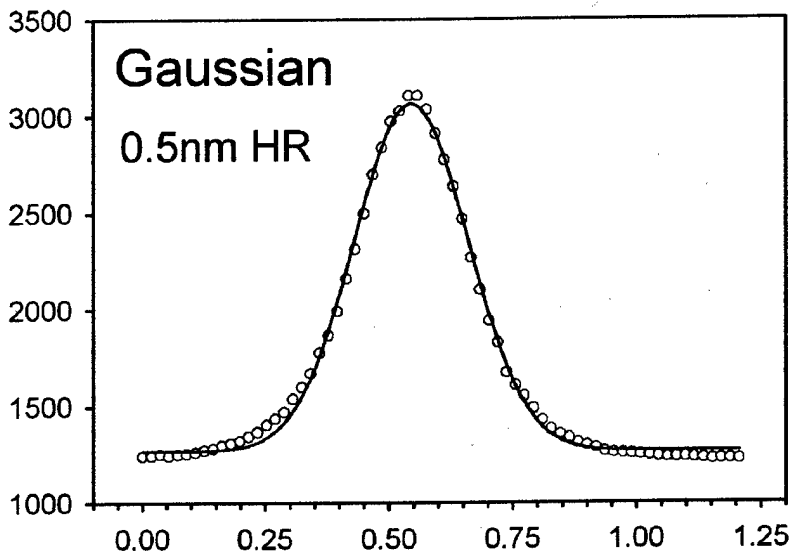


Figure 2. Current distribution in the probe used for data in Fig. 1. FWHM is 0.256 nm. Current in the probe was 51 pA.

is a Gaussian fit to the distribution, which quite accurately describes it. The full width at half maximum of the distribution is ~ 0.25 nm. Convolution of this distribution with a chemically sharp interface (Heaviside step function) broadens it to ~ 0.5 nm, or twice the FWHM. This width is smaller, by about a factor of 2, than the sharpest chemical distributions shown on the composition plot of Fig. 1. Thus they are

real distributions. These chemical distributions affect the dielectric constant of the total stack, and are important information useful for the research being done to develop a replacement for SiO_2 in gate dielectrics for CMOS devices. They have not been measured before this work.

The ultimate electromechanical stability of STEM electron microscopes like this one is

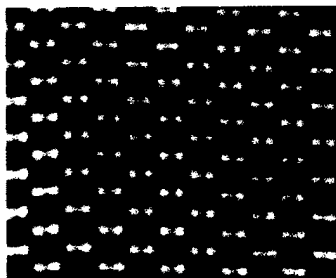


Figure 3. Si Dumbbells

verified by imaging what are called silicon "dumbbells". An image of Si dumbbells taken with this microscope is shown in Fig. 3. Here, the beam is along the [011] direction of the Si specimen (i.e. that is the projection direction of the image) and the dumbbell axis projected length along the [200] Si direction in the plane of the image is 0.14 nm. This type of image is acquired with a smaller probe containing less current than the one shown in Fig. 2. It does demonstrate the excellent electromechanical stability of the microscope system, which includes all its peripherals and its environment. The environmental stabilization we provided is necessary to

achieve this level of performance. There are perhaps 10 other STEM microscopes throughout the world with this performance, and 3 that I can recall in the US. There are many other microscopes of this same type installed in sites that do not permit this level of performance. This was a lengthy and at times difficult installation, but the results do certainly justify the effort. This microscope should produce many materials research results in the future.

Its performance will not be significantly bettered until the next generation of STEM microscopes is built using multipole objective lens "correctors", which will allow the spherical aberration coefficients of their probe forming lenses to be reduced to arbitrarily small values, thus enabling improved performance. A few existing STEM microscopes have been retrofitted with the first prototype correctors to be produced, but their performance is restricted by their environment. The first of the new generation STEM microscopes from JEOL is scheduled to be installed in May of 2004, at Oak Ridge National Laboratory. A new building is being constructed for it, to ensure adequate environmental stability. The cost of the installation is \$3,000,000 for the microscope plus the building's cost. The total will be four or more times the total cost of the present microscope system. In this context the present microscope is an excellent value for the funds and effort expended, and will remain very competitive for 5 to 10 years, particularly in a university environment.

On behalf of my colleagues and myself, we thank you and the AFOSR for funding this project.

Very truly yours,

R. W. Carpenter, Professor and Project Director.

xc: Co-investigators:
Prof. Wm. Petuskey
Prof. D. J. Smith