Highly charged ion (HCI) modified tunnel junctions

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Abstract. The neutralization energy carried by highly charged ions (HCIs) provides an alternative method for localizing energy on a target's surface, producing features and modifying surfaces with fluences and kinetic energy damage that are negligible compared to singly ionized atoms. Since each HCI can deposit an enormous amount of energy into a small volume of the surface (e.g., Xe^{44+} delivers 51 keV of neutralization energy per HCI), each individual HCI's interaction with the target can produce a nanoscale feature. Many studies of HCI-surface features have characterized some basic principles of this unique ion-surface interaction, but the activity reported here has been focused on studying ensembles of HCI features in ultra-thin insulating films by fabricating multi-layer tunnel junction devices. The ultra-thin insulating barriers allow current to flow by tunneling, providing a very sensitive means of detecting changes in the barrier due to highly charged ion irradiation and, conversely, HCI modification provides a method of finely tuning the transparency of the tunnel junctions that spans several orders of magnitude for devices produced from a single process recipe. Systematic variation of junction bias, temperature, magnetic field and other parameters provides determination of the transport mechanism, defect densities, and magnetic properties of these nano-features and this novel approach to device fabrication.

Keywords: magnetic tunnel junction, highly charged ions, electron beam ion trap (EBIT)
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INTRODUCTION

For more than 50 years, the use of an atom or ion's kinetic energy to modify materials, generally through "sputtering," has been thoroughly documented[1] and reasonably well understood[2]. If one is interested in producing nano-scale surface modifications from a *single* incident ion, one generally needs to use kinetic energies in the hundreds of keV or greater, which produces extended tracks of dislocations and vacancies through the bulk of the target. To produce features localized at the target-vacuum interface using kinetic energy, it is therefore necessary to use a large ensemble of low energy ions that are collectively focused to scribe the feature of interest, e.g., focused ion beams (FIB).

Highly charged ions (HCIs) provide a fundamentally different approach to target modification by providing an alternative supply of energy: the ion's neutralization energy. As additional electrons are removed from an ion, the total neutralization energy accumulates rapidly. For the HCIs used in the work described here, neutralization energies are much more significant per unit volume at the surface (neutralization energy for Xe^{44+} is > 51 keV deposited into order 1 nm^3) than the kinetic stopping energies (≈ 3 keV/nm at 360 keV; max stopping power for Xe is ≈ 27 keV/nm at 700 MeV). Since the HCI's neutralization energy is completely independent of its velocity, *all* this energy is deposited at the target-vacuum interface, and can be channeled into nano-features.

This report reviews recent progress made at the National Institute of Standards and Technology (NIST) Electron Beam Ion Trap (EBIT) facility studying the efficacy of modifying ultra-thin electron tunnel barriers using HCIs. Many years of the fundamental studies of HCI-target interactions performed by several groups worldwide enable this effort; due to space limitations, only a few representative citations throughout will be provided to allow the avid reader to explore the field[3-10]. Generally, previous work shows that metals have weak potential (neutralization) energy sputtering (PES), good insulators have dramatic PES, and complex materials in between have complex energy dissipation chains that do not always lead to strong PES, but typically have large secondary electron yields. Since

the work here is a foray into technological implementation of HCIs while seeking to verify fundamentals, we have exclusively used aluminum oxide as the ion target, previously shown to have very strong PES[11].

SAMPLES

All tunnel junctions (TJs) described have the same basic structure and fabrication method in common, see Ref. [12] for more details. The TJs in this study are nearly 2D devices of sizes ≈ 50 µm x 50 µm composed of two metallic multi-layers with a total thickness ≈ 30 nm separated by an aluminum oxide tunnel barrier typically between 1 nm and 2 nm in thickness, depending on the study. The devices are grown through a set of shadow masks that have two crossing lines, both defining the device's geometry, and also forming four connecting electrodes that provide for four-point electrical probing. An experiment involving HCI irradiation proceeds as follows: 1) oxidized, blank Si chips cleaned *ex situ* by standard techniques are load locked into vacuum, 2) the substrates are re-cleaned *in situ* using oxygen plasma, 3) the chips are then transferred *in situ* to an electron-beam deposition system where the lower metallic multi-layers and connecting electrodes are deposited (4 devices per chip), 4) an ultra-thin aluminum layer is deposited over the entire chip, 5) the chip is exposed to an oxygen plasma for a time determined to oxidize the aluminum layer completely with minimal over-oxidation, 6) chips are held in ultra-high vacuum (UHV) until a full batch is ready, 7) three devices per chip are exposed to a controlled fluence of HCIs, leaving one undosed control device per chip, 8) the upper metallic multilayer and electrodes are deposited immediately following HCI irradiation, and, finally, 9) the chips are removed from vacuum for measurement. (Note – steps 2-8 all occur within a contiguous UHV vacuum system without removal. A simplified device schematic is inset in Figure 1.)

RESISTANCE-AREA PRODUCT AND HCI ADJUSTMENT

The principal technological motivation for this work is to demonstrate a method of producing tunnel junctions spanning a wide range of resistances using a single process recipe. Achieving variable conductance in a single recipe is of great interest to the magnetic sensor (hard drive) and magnetic random access memory (MRAM) industries, where junction areas tend to change between device generations as magnetic bits are shrunk. Since any given layer structure has a fixed resistance-area (RA) product

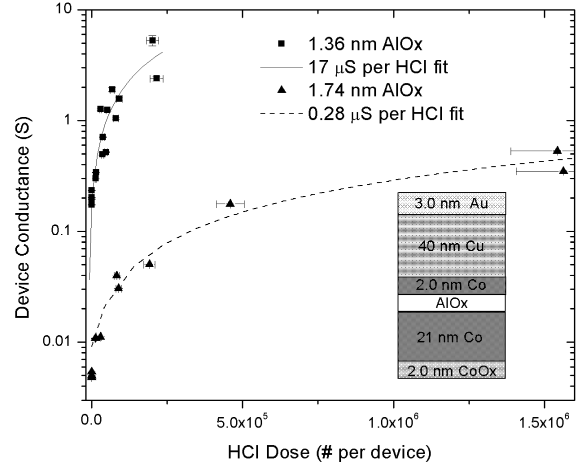

Figure 1. Conductance as a function of HCI dose for two sets of magnetic tunnel junction samples, one with a 1.36 nm tunnel barrier and the other with a 1.74 nm tunnel barrier. The thinner barrier system displays a much stronger conductance increase due to the HCI irradiation, i.e., 17 µS per HCI compared with 0.28 µS per HCI for the thicker barriers. All samples are otherwise identical, processed in the same batch, and irradiated on the same day. Measurements were performed at room temperature, at < 10 mV bias, and with no externally applied fields; error bars represent +/- 1 standard uncertainty from both statistical and measurement errors. Inset – simplified device layer structure.

(two-dimensional resistivity), a change in device area correlates to a change in its resistance. Since product engineering and performance are optimized when the resistance remains constant, each product generation requires an adjustment to the RA product, which conventionally requires a revision of the fabrication process. Using HCI irradiation, a reduction in the RA product is instead accomplished by step 7 above; when individual HCIs neutralize on the tunnel barrier and create nanometer sized regions of increased conductance that accumulate and dominate the device's transport. The device's impedance is then determined by the total density of the HCI irradiation. HCI adjustment of the RA product using a single process recipe is demonstrated spanning a range of more than three orders of magnitude[13], and, in principle, can span many more.

As an example, device conductance as a function of HCI irradiation for two tunnel barrier thicknesses in otherwise identical samples is shown in Figure 1. While both tunnel barrier thicknesses exhibit enhanced conductance, thinner barriers experience a larger conductance increase per HCI. This large difference in rate of conductance change between the two systems suggests the transport is still dominated by tunneling, since a fixed incremental reduction in the barrier thickness has a much smaller absolute effect on a thicker barrier. Current vs. bias measurements

confirm this assessment, and Simmons model[14] fits of these data are consistent with the expected thickness and potential barrier heights of the TJs. Resistance as a function of temperature data allow us to compare the density of "defect" states that promote "nonlinear hopping" transport across the barrier, which is found to be similar in irradiated and un-irradiated devices, about 20%. These results taken together indicate that the HCI processing reduces the effective tunnel barrier thickness in the ion's impact zone, increasing the tunnel probability while retaining the initial quality of the barrier and without creating conductive pinholes, discussed further in Ref. [13].

RECENT ACTIVITY

Having demonstrated HCI enhanced conductance in TJs, two intertwined questions are being pursued: 1) can the HCI-reduction approach be employed in a system which more closely mirrors devices of industrial interest, e.g., devices that exhibit magneto-resistance, and 2) how robust is the process and what properties of the device, specifically the tunnel barrier, are required for HCI-mediated RA reduction to be successful? To address these questions, many variations of tunneling devices are under study, but principally involve devices that exhibit magneto-resistance (MR), i.e. change in the device resistance due to the application of an external magnetic field. Some of the parameters varied include: the tunnel barrier thickness, the different magnetic materials, presence of antiferromagnets (AFMs), oxidation process parameters that may leave under-oxidized aluminum or oxidize beyond the aluminum into the underlying magnetic layer, etc. For the purposes of this report, only a couple will be mentioned briefly.

In order for a TJ to exhibit MR, a) magnetic layers that spin polarize the electric current must be immediately adjacent on either side of the tunnel barrier, and b) the coercivities of these two layers must be substantially different so that the magnetic moments of the two layers can be independently oriented, e.g., aligned oppositely. (The MR quantity $\Delta R/R_{min}$ of a device is proportional to the dot product of the magnetic moments of these two magnetic layers.) In this work, cobalt and permendur (CoFe) are both used to form magnetic layers and coupling one layer to an AFM (cobalt oxide) is used to create a large difference in the coercivity of the layers. Although cobalt oxide (CoOx) is used due to the technical compatibility with our deposition system, it has drawbacks: the Néel temperature is below 0 ºC (magnetic measurements must be performed cold, i.e., liquid nitrogen temperature) and it is an insulating material, which requires that the CoOx be at the very

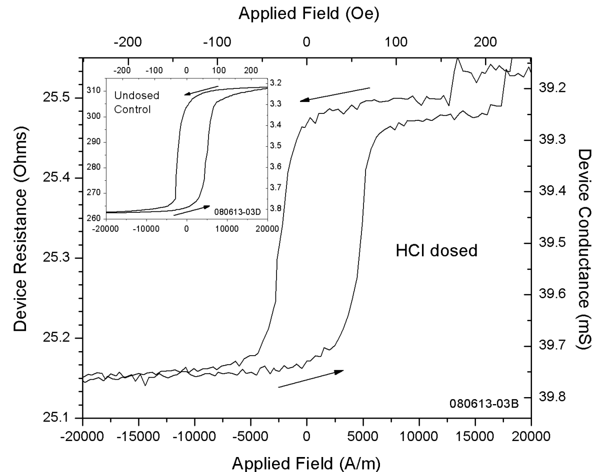

Figure 2. Conductance as a function of magnetic field (arrows indicate field sweep direction) for an irradiated and corresponding control (inset) magnetic tunnel junctions (MTJs) from the thicker (1.74 nm) barrier system shown in Figure 1. While the irradiated sample's qualitative response is the same as the control, the magnitude of the magneto-conductance remains the same (≈ 0.7 mS), indicating the HCI induced current paths are spin insensitive, unlike observations of thinner barrier MTJs. Measurements were performed in liquid nitrogen, at < 10 mV bias, and one standard deviation in the uncertainty of resistance is ≈ 20 mΩ and in field is ≈ 80 A/m (1 Oe) from measurement sources.

top or very bottom of the device layer structure. After many variations, placing CoOx at the extreme bottom and coupling it to a moderately thick conducting cobalt layer produced the best MR and the most reliable devices. In some cases, the HCI-enhanced conductance is found to exhibit MR of about 6 %, while in some other cases, the HCI-channels show little or no MR. As an example, MR data for two of the 1.74 nm AlOx devices shown in Fig. 1 (2.0 CoOx/20.0 Co/1.0 Co/1.74 AlOx/10.0 Co/40.0 Cu/3.0 Au – thicknesses in nm) are shown in Fig. 2, (other samples in Fig. 1 have an identical recipe except for the 1.36 nm AlOx). The MR data in the main panel of Figure 2 was taken on an HCI dosed device, while the inset is from an un-irradiated, identical control device. Surprisingly, these thicker barriers appear to maintain the MR poorly compared to other thinner barriers, the conductance change in Figure 2 is equal to the conductance change in the base tunnel junction, indicating little or no MR contribution from the HCI-enhanced conductance. Even though the total MR is diminished, the quality of tunnel transport (from I-V curves) does not appear to be worse, implying a spin scattering mechanism that is either elastic or not within the tunnel barrier in the HCI reduced regions. As a possible explanation, the HCI irradiation may produce aluminum rich areas at the HCI impact sites that

reduce the electron polarization, thereby quenching the MR in those channels. Other device recipes using thinner barriers maintain the MR better, possibly due to an inadvertently higher oxygen concentration that allows unbound oxygen to re-oxidize the aluminum rich areas formed by HCI impact.

SUMMARY

The neutralization energy of HCIs offers a promising alternative to modification of materials from the traditional use of an ion's kinetic energy. In the work described here, HCIs are used to systematically increase the transparency of tunnel junction devices without any other variation in the process recipe. HCI irradiation can be compatible with devices that exhibit meaningful magneto-resistance using many different process recipes. Ongoing work is aimed at identifying processing parameters that simultaneously optimize desirable HCI processing sensitivity and retain MR.

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