Experimental and theoretical studies of the energy-loss straggling of H and He ion beams in HfO$_2$ films


$^1$Departament de Física Aplicada, Universitat d’Alacant, Apartat 99, 03080 Alacant, Spain
$^2$Instituto de Física, Universidade Federal do Rio Grande do Sul, Av. Bento Gonçalves 9500, 91501-970 Porto Alegre, RS, Brazil
$^3$Departamento de Física - CIQyN, Universidad de Murcia, Apartado 4021, 30080 Murcia, Spain
$^4$División Colisiones Atómicas, Centro Atómico Bariloche, 8400 San Carlos de Bariloche, Argentina
$^5$Centro Brasileiro de Pesquisas Físicas, 22290, Rio de Janeiro, RJ, Brazil

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Abstract. We report an experimental-theoretical study of the energy-loss straggling of protons and alpha particles in HfO$_2$ films. In the case of H ions the experiments were performed in the energy range 40–1750 keV. For the lower energy interval (40–250 keV) we have used the medium energy ion scattering (MEIS) technique with a resolution of $\Delta E/E \sim 4 \times 10^{-3}$, while for the higher energies the Rutherford backscattering technique (RBS) was employed with an overall resolution of 7 keV. Concerning the He ions the straggling study has covered an energy range between 250 and 3000 keV by using RBS measurements, which in this case had a resolution better than 10 keV. The theoretical calculations were done in the framework of the dielectric formalism using the MELF-GOS model to obtain a proper description of the energy loss function (ELF) of the HfO$_2$ target. It is shown that for both projectiles the experimental data and the theoretical predictions for the energy-loss straggling display a very good agreement.

PACS. 34.50.Bw Energy loss and stopping power – 77.22.-d Dielectric properties of solids and liquids

1 Introduction

When a fast light ion moves through matter, its initial energy is lost in the target due mainly to interactions with the electrons of the material. As these interactions are of statistical nature, during the motion of the fast projectile inside the target there are fluctuations in the individual energy-transfer values, as well as in the number of interactions taking place. Therefore a spread in the energy-loss distribution appears, which gives rise to the so called energy-loss straggling.

A good control of the energy deposited by an energetic projectile is essential for achieving a reasonable interpretation in ion beam analysis or a satisfactory result in ion beam modification of materials. The main parameters that quantify the energy deposition are the electronic stopping power (which is the mean energy deposited per unit path length) and the energy-loss straggling (which represents the mean square deviation of the energy loss distribution per unit path length) [1]. Both magnitudes are widely studied experimentally as well as theoretically, although results for the latter are much scarcer than for the former, in particular for the case of compound targets.

Therefore, further work on the energy-loss straggling in elemental and, in particular, compound targets is necessary. These studies are very important in the microelectronics industry, because when dealing with thin films the undesirable broadening of the deposited energy (due to the straggling) may significantly affect the profile of the implanted dopant [2].

At low incident H and He ion velocity, the energy-loss straggling is only affected by the target valence electrons, while at higher projectile velocities the more tightly bound electrons in the target atomic core also contribute [3,4]. Therefore for compound materials the energy-loss straggling at low and intermediate projectile velocities is affected by aggregation (chemical and phase) effects while at higher energies the participation of the target atomic core electrons increases, which behave almost as when they belong to isolated atoms.

Assuming that all the target electrons contribute to the energy loss, Bohr [5] provided a simple expression for the value of the energy-loss straggling in the case of an elemental target:

$$\Omega_B^2 = 4\pi Z^2_1 e^4 Z_2 N,$$ (1)
where $Z_1$ and $Z_2$ are the projectile and target atomic numbers, respectively, $e$ is the electronic charge and $N$ is the target atomic density. Equation (1) is referred to as the Bohr straggling and is frequently used to estimate the corresponding energy-loss straggling value for the case of high projectile velocities. The value of $\sigma_{B}^2$ can be also calculated straightforwardly for compound materials through Bragg’s additivity rule [6] applied to their elemental constituents.

There is some available information in the literature on the straggling for metals or, more generally, conductors. However in the case of insulators the measurements are more complicated and the experimental information is scarce [7]. The major experimental difficulties concern the smoothness and homogeneities of the film under study, as well as target charging effects.

Due to its high dielectric constant, wide energy bandgap energy [8], and good thermal stability [9], HfO$_2$ films play a strategical role in the microelectronics industry because they already start to replace the SiO$_2$ films in metal oxide field effect transistors (MOSFET) allowing further miniaturization [10]. In fact, for this compound there is a lack of information about the straggling and even on the stopping power for light ions, which should be of great utility for the industry oriented research, as well as for basic studies [11].

In this paper we present energy-loss straggling measurements and calculations for H and He ion beams in HfO$_2$ films. For the H beams the experiment was done in the energy range from 40 keV up to 1750 keV, while for the He ions, the investigated energy range was between 250 and 3000 keV. The experimental results are compared with theoretical calculations based on a proper description of the dielectric properties of the target [12,13] adapted to the present case of a HfO$_2$ target.

The rest of this work is organized as follows. In Section 2 the experimental procedure and details are described, while the theoretical model is outlined in Section 3. The comparison of experimental and theoretical results is shown in Section 4, and finally a summary is drawn in Section 5.

2 Experimental procedure

2.1 Sample preparation

The HfO$_2$ films were grown on a Si (100) substrate by radio frequency (rf) magnetron sputtering (150 W) using a HfO$_2$ target with a nominal purity of 99.95% and O$_2$/Ar gas mixture as sputtering gas. The sputtering system was evacuated to $8.0 \times 10^{-8}$ torr by a turbo molecular pump backed by a mechanical pump before the deposition. The total work pressure was 5.7 mtorr during the deposition with an Ar gas flow of 19.6 sccm and an O$_2$/Ar ratio flow of 0.35 was used. The deposition rate (3.3 nm/min) was checked by the analysis of low-angle X-ray reflectivity scan on one of the HfO$_2$ films, and the thicknesses ($t = 17, 32, 63, 72, 91$ and $1450 \text{ nm}$) of the HfO$_2$ films were controlled using the deposition time and after check by the X-ray reflectivity technique. A Phillips X-Pert $\theta$–$\theta$ diffractometer employing Cu $K\alpha$ radiation was used to obtain the low and high angle diffraction scans. The surface was observed by atomic force microscopy (AFM) using a NanoScope-IIIA from Digital Instruments. The mean square roughness of the films was on average less than 5% of the total film thickness and that of the Si substrate was 0.3 nm. The stoichiometry of the films was checked using the Rutherford backscattering technique (RBS) and the fitting on the experimental results has confirmed that in fact we are in presence of stoichiometric HfO$_2$ films. Also we have used a very thin film ($t = 5 \text{ nm}$, with mean roughness less than 0.5 nm) provided by IBM, Yorktown, USA, in order to perform the measurements with very low energy H beams.

2.2 Experiments and measurements

For the case of H$^+$ projectiles we have used the medium energy ion scattering (MEIS) technique for the lower energies (from 40 up to 250 keV), with a resolution $\Delta E/E \sim 4 \times 10^{-3}$. For higher energies we have used the standard Rutherford backscattering (RBS) technique. With this aim we had two detectors with an overall resolution of 7 keV situated symmetrically at $\pm 165^\circ$ with respect to the beam direction. For each analyzed energy, a suitable set of samples with different thicknesses was used. In each case RBS spectra were taken at $\theta_1 = 0^\circ$, $30^\circ$ and $45^\circ$ with respect to the normal of the sample. Since at non-normal incidence (e.g. under a target tilt different from $\theta_1 = 0^\circ$) a non-symmetrical detector geometry is mandatory, then two different spectra are obtained for each energy, each one corresponding to each detector situated at different angle with respect to the beam direction. In order to obtain the energy-loss straggling we have analyzed only the Hf component of the film, since the O part laid on top of the Si spectra corresponding to the Si wafer and the straggling analysis would be very hard. Therefore, for each energy we had at least six spectra. From them, as will be described in the next sub-section, the corresponding straggling was extracted and an average value was obtained.

The determination of the energy-loss straggling for He$^+$ projectiles in the 250–3000 keV energy interval was done using only the RBS technique with an overall resolution better than 10 keV. The experimental set-up and procedure was the same as described for the H$^+$ case and consequently again for each energy we had at least six spectra from where the individual straggling was extracted and an average value was obtained.

Figure 1 shows a typical RBS spectrum of the Hf component of the HfO$_2$ film for the case of 600 keV He$^+$ incident with $\theta_1 = 45^\circ$ with respect to the normal of the 72 nm sample; the red continuous line represents the fitting to the experimental data, which are depicted by symbols. As it may be noted, by comparing the front and backside of the spectrum a straggling produced in the sample is clearly observed. The inset in Figure 1 illustrates the whole corresponding RBS spectrum.