Available online at www.sciencedirect.com
ScienceDirect
Journal of Non-Crystalline Solids 354 (2008) 3025–3033
JOURNAL OF NON-CRYSTALLINE SOLIDS
www.elsevier.com/locate/jnoncrysol

Electronic structure and charge transport properties of amorphous Ta_2O_5 films

V.A. Shvets [a], V.Sh. Aliev [a], D.V. Gritsenko [a], S.S. Shaimeev [a], E.V. Fedosenko [a], S.V. Rykhlitski [a], V.V. Atuchin [a], V.A. Gritsenko [a], V.M. Tapilin [b], H. Wong [c,*]

[a] *Institute of Semiconductor Physics, SB RAS, Novosibirsk 630090, Russia*
[b] *Institute of Catalysis, SB RAS, Novosibirsk 630090, Russia*
[c] *Department of Electronic Engineering, City University of Hong Kong, Tat Chee Avenue, Kowloon, Hong Kong*

Received 6 September 2007; received in revised form 27 November 2007
Available online 4 March 2008

Abstract

Amorphous Ta_2O_5 films were deposited by sputtering Ta onto silicon substrates with reactive ion beam. Electron energy loss spectroscopy measurements on the film found that the plasma oscillation energy is 23.1 eV. The refractive index and the extinction coefficient were measured with spectroscopic ellipsometry over the spectral range of 1.9–4.9 eV. The optical band gap is found to be 4.2 ± 0.05 eV. The valence band consists of three bands separated by ionic gaps. The values of electron effective masses were estimated with DFT quantum-chemical calculation. Experiments on injection of minority carriers from silicon into oxide were also conducted and we found that the electron component of conduction current governed by the electron current in the amorphous Ta_2O_5.

PACS: 68.49.Uv; 71.15.Mb; 71.20.−b; 72.20.−i; 78.20.−e

Keywords: Band structure; Dielectric properties, relaxation, electric modulus; Ellipsometry; Atomic force and scanning tunneling microscopy; Ab initio; XPS

1. Introduction

As the amorphous tantalum pentaoxide (Ta_2O_5) film possess high thermodynamic stability and with large refractive index of 2.1, it was used as antireflection coating for silicon solar cells and used in the multilayer interference filters and other optical devices [1]. Recent intensive investigations focus on the applications of this film in dynamic random access (DRAM) capacitors [2–4]. Meanwhile, the reduction of gate dielectric thickness in metal-oxide-semiconductor (MOS) structures has prompted the search for higher dielectric constant (high-k) oxide materials to replace the traditional SiO_2 ($k = 3.9$) and Si_3N_4 ($k = 7$) [5,6]. Among the high-k materials, Ta_2O_5 films received considerable attention because of its high dielectric constant ($k = 25$). Besides this, high-k materials may be used as blocking dielectric in a gate stack of flash memory devices [7]. In this article, we investigate physical and electrical properties of amorphous Ta_2O_5 films prepared by ion-beam sputtering deposition (IBSD) method. We shall focus on the electronic structure and the electrical characteristics of this oxide. Polycrystalline Ta_2O_5 films are used in DRAM for better charge storage and the current leakage is crucial for this application. It is well known that the leakage current is governed Fowler–Nordheim (FN) tunnel mechanism and the effective mass of carrier is one of the most important parameters. In this work, we determined, for the first time, the carrier effective masses in Ta_2O_5 using quantum-chemical simulation. On the other hand, the charge transport in Ta_2O_5 films can be contributed by either electrons or holes. To have a better understanding of the charge transport properties, separation of the electron and hole currents are also important. The work also

* Corresponding author. Tel.: +852 27887722; fax: +852 27887791.
E-mail address: heiwong@ieee.org (H. Wong).

0022-3093/$ - see front matter © 2008 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnoncrysol.2007.12.013

conducted experiment to separate these currents. In addition, the optical constant dispersions of Ta_2O_5 films are also studied, for the first time, using spectroscopic ellipsometry.

2. Experimental details

The tantalum pentaoxide (Ta_2O_5) films were prepared by ion-beam sputtering deposition system illustrated in Fig. 1. The deposition chamber was initially evacuated by turbo molecular pump to a residual pressure of 2×10^{-5} Pa. This chamber is equipped by Kaufmann ion source (1) with ion-beam diameter of 6 cm. The target used for the deposition was tantalum with 99.95% purity (2). Tantalum disc sputtering was produced by Ar^+ beam with ion energy of 1200 eV and current density of 1.2 mA/cm^2. During the film deposition the substrate temperature was maintained at lower than 70 °C. The tantalum atoms reacted with oxygen atoms from the oxygen source (3) and deposition on the substrate (4). The film thickness is monitored with a quartz crystal microbalance (5). The system equipped with additional lock-chamber (6) with RF plasma cleaning system (7). During the deposition the pressure was changed to about 1.5×10^{-2} with the inlet of gas mixture with Ar/O_2 ratio of 2. The (1 0 0) Si wafers with resistivity of about 5–7 Ω cm were used as the substrates. Just before the insertion of the substrate into the chamber, the native oxide of the substrate was removed HF etching. In several experiments the substrates were subjected to supplementary surface treatment in lock-chamber by low energy Ar^+ ion bombardment.

The crystallographic properties of the Ta_2O_5 films were controlled by a reflection high-energy electron diffraction (RHEED) at an electron accelerating voltage of 50 kV. A charge neutralization flood gun was utilized to eliminate charging effects. Top surface chemical composition and electronic parameters of the oxide films were determined by Riber LAS 3000 X-ray photoelectron spectroscopy (XPS) with monochromatic Al K_α ($hv = 1486.6$ eV) excitation source. Electron energy loss spectra (EELS) were measured using the same spectrometer. Bandgap of the tantalum pentaoxide film was measured by EELS using monochromatic electron beam with energy of 200 eV.

The surface morphology of the films was studied with a Solver P47 H atomic force microscopy (AFM). This microscope provides a lateral and depth resolutions of 15 nm and 0.05 nm, respectively. The AFM images were generated in resonance frequency of 150 kHz in semi-contact mode with platinated cantilever.

Spectroscopic ellipsometry measurements were carried out using an instrument operating at a balance configuration and Xe lamp was used as a light source. The spectrum ranging from 1.9 to 4.9 eV and has a resolution of 0.05 eV. The incident angle was fixed at 70.6°. Ellipsometric surface mapping was performed on LEF-8 MICROSCAN single-wavelength ($\lambda = 632.8$ nm) instrument equipped with specially designed micro-objective and stepper-motor-driven scanning stage. In-plane resolution of the map was determined mainly by the size of the focused laser beam (ellipti-

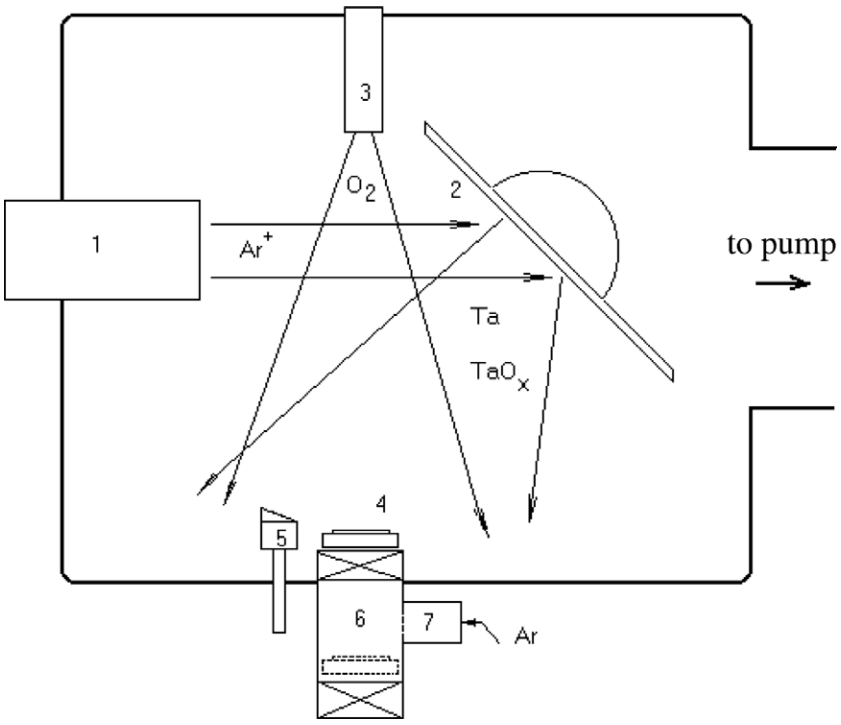

Fig. 1. Set up for reactive ion deposition. The components are: 1 – ion source, 2 – tantalum target, 3 – oxygen source, 4 – substrate, 5 – quartz crystal microbalance, 6 – lock-chamber, 7 – high-frequency input.