Study on absorbance and laser damage threshold of HfO_2 films prepared by ion-assisted reaction deposition

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Using a new kind of EH1000 ion source, hafnium dioxide (HfO_2) films are deposited with different deposition techniques and different conditions. The absorbance and the laser damage threshold of these films have been measured and studied. By comparing these characteristics, one can conclude that under right conditions, such as high partial pressure of oxygen and right kind of ion source, the ion-assisted reaction deposition can prepare HfO_2 films with higher laser induced damage threshold.

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For its relatively high damage threshold and good thermal and mechanical stability, hafnium dioxide (HfO_2) is one of the most commonly used high-index materials to realize laser damage resistance applications[1−6]. However, the problem of preparation of higher laser damage threshold HfO_2 films has not yet been solved. The traditional way is to evaporate oxide hafnium which is the easier way to proceed, but creates nodular defects in the coatings which can limit the laser damage threshold[4]. Preparation of HfO_2 films using hafnium as starting materials can reduce these defects, but the problems of whether and how to use ion-assisted reaction deposition during this process have not yet been solved. The disadvantages introduced by ion or ion source have been investigated[3−6]. These works enlighten us on preparation of HfO_2 films by ion-assisted reaction deposition with few disadvantages. In our work, HfO_2 thin films with different deposition techniques and deposition conditions have been prepared using a new kind of end-hall ion source, and laser damage threshold of these films has been studied.

The vacuum system was pumped by means of turbomolecular pump. HfO_2 films have been deposited on BK7 glass substrate (ϕ30×4 mm) in ZZSX-800F vacuum coating system equipped with an EH1000 ion source. HfO_2 films have the same thickness of 340 ± 10 nm monitored by optical control. The substrates were cleaned ultrasonically in alcoholic solution and their temperature values were 300 °C when samples were preparing. The base vacuum of all depositions was 1.0×10^{-3} Pa, while during deposition the chamber pressure changed because of the presence of argon gas and oxygen gas. The argon gas was used to protect the ion source cathode, and the oxygen gas was used to provide ions that bombard thin films during film deposition process. The EH1000 ion source has the advantages of other end-hall ion sources, such as gridless, wide-angle ion beam and low ion energy with high current[5]. Furthermore, as a new kind of end-hall ion source, it has its own characteristic: no filament. Samples B and C were deposited with same ion current, chamber pressure and different ion energy, however samples C and D were deposited with same ion energy and different ion current, chamber pressure. The deposition parameters are given in Table 1, where I_b means the emission current.

Surface thermal lensing method has been used to characterize the weak absorption of samples[7]. To this end, a single-mode, continuous wave (CW) YAG 1064-nm laser with a beam diameter of 100 μm has been used as the pump source. The accuracy of the measurement is 1×10^{-6}. Experimental apparatus was the same as that in Ref. [7]. Figure 1 shows the absorbance of samples.

Damage testing was performed in the "1-on-1" regime[8,9]. The experimental setup used here was the same as that in Ref. [8]. The Q-switched Nd:YAG laser produced a TEM_{00} mode with a 12-ns pulse width. The laser-induced-damage threshold (LIDT) was defined as the incident pulse's energy density when the damage occurs at 0% probability (unit: J/cm^2). Figure 2 shows the values of LIDT of the samples.

Damage morphologies of samples were obtained by Leica DMRXE micropolariscope. Figure 3 shows the damage morphologies of samples when the irradiation energy was the double of each sample's LIDT.

Comparing absorbance and LIDT of samples A and D from Figs. 1 and 2, one can conclude that the absorbance and the damage threshold of the tested HfO_2 films depend on the film deposition technique. Sample A grown

Table 1. Deposition Parameters Used to Obtain the HfO_2 Films

Sample	Starting Material	I_b (mA)	O_2 Pressure (Pa)	Ion Energy (eV)	Ion Current Density (μA/cm^2)
A	HfO_2	160	7.5×10^{-3}	—	—
B	Hf	200	2.3×10^{-2}	84	140
C	Hf	200	2.3×10^{-2}	60	140
D	Hf	200	3.4×10^{-2}	60	220

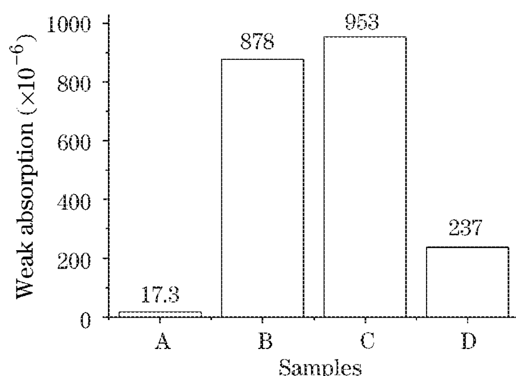

Fig. 1. Absorbance of the samples.

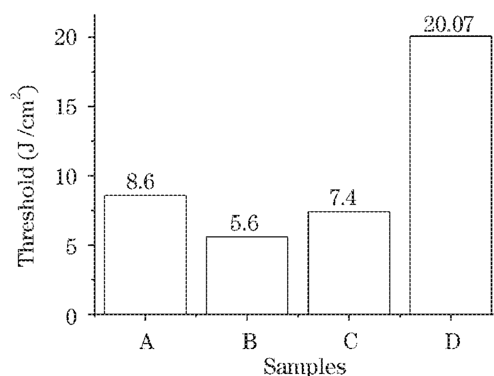

Fig. 2. Laser damage threshold of the samples.

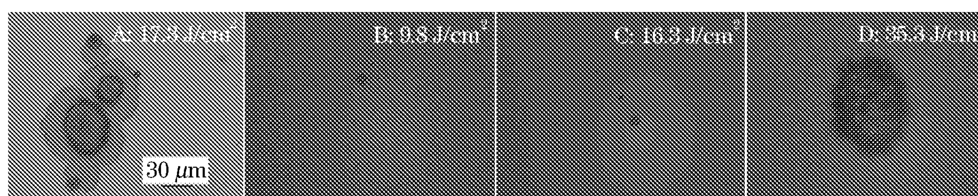

Fig. 3. Damage morphologies of samples.

without ion assistance has the lower absorbance using HfO_2 as starting material, and sample D grown with ion assistance has the higher threshold using hafnium as starting material.

It is well known that one film with high LIDT must have low absorbance, so it is interesting to note that the absorbance of sample D (237×10^{-6}) is more higher than that of A (17.3×10^{-6}), but the threshold of sample D (20.07 J/cm^2) is more higher than that of A (8.6 J/cm^2). Let us try investigating the reason.

From damage morphologies of Fig. 3, one can conclude that damage is mainly due to absorption at defect sites, especially for sample A. One defect localized in film is an absorbing inclusion that can be heated by laser radiation up to a high temperature. During being heated, film creates free electrons those lead to additional absorption, and the temperature of defect surrounding can be very high. So the thermal explosion takes place. Using HfO_2 as starting material is a traditional way to produce HfO_2 films, but it creates nodular defects in the films, and the reaction deposition using hafnium as starting material can reduce the nodular defects[4]. It is the nodular defects that limit the LIDT of sample A greatly. Some works reveal that during ion-assisted deposition process, the filament of ion source will introduce impurity defects into films[10]. Because the EH1000 ion source is filamentless and gridless, here, it is no need to discuss impurity defects introduced by ion source.

About the absorbance, it is worth noting that the pump source in weak absorption experiment is a CW YAG 1064-nm laser while in laser damage experiment the induced laser source is a Q-switched pulsed Nd:YAG 1064-nm laser. The nodular defects may be sensitive to pulsed laser but not be sensitive to CW laser because the dimension of nodular defects usually is smaller than laser wavelength (1064 nm). So the nodular defects have great influence on the LIDT of sample, but it cannot have enough influence on the photo-thermal signal. Comparing absorbances of sample C and D, we can find the metallic characteristics of films have great influence on the absorbance when measured using CW laser which will be discussed later. Here, one can suppose that the absorbance measured by CW laser is the reflection of intrinsic absorption especially metallic characteristics—the main problem of reactive deposited HfO_2 films[4]. So, the absorbance of sample D with metallic characteristics is higher than sample A.

Comparing absorbance and LIDT of samples B, C, and D from Figs. 1 and 2, one can conclude that the absorbance and the damage threshold of the tested HfO_2 films depend on the film deposition conditions. Though three samples are all prepared by ion-assisted reactive deposition, among them, sample C has the highest absorbance, and sample B has the lowest LIDT, while sample C has the highest LIDT.

Samples B and C deposited at the same partial pressure of oxygen (2.3×10^{-2} Pa) have the higher absorbance (878 and 953×10^{-6} respectively), while the absorbance of sample D deposited at the partial pressure of oxygen about 3.4×10^{-2} Pa is 237×10^{-6}. High partial pressure of oxygen reduces metallic characteristics of HfO_2 films[4]. Associating it with the absorbance of films, one can deduce that the absorbance measured by CW laser is the reflection of intrinsic absorption, especially metallic characteristics. Because samples B, C, and D were all deposited by ion-assisted reaction, the defects densities of three samples were at the same order. Under this condition, intrinsic absorption is the main factor that limits the LIDT of films. So, sample D deposited under high partial pressure of oxygen with little metallic characteristics has the highest LIDT.

In summary, using the new kind of EH1000 ion source,

HfO_2 films were deposited with different deposition techniques and different conditions. By comparing the absorbance and LIDT of samples, one can conclude that:

1) The absorbance and the damage threshold of the tested HfO_2 films depend on the film deposition technique and conditions.

2) Reducing the nodular defects by using metal hafnium as starting material, reducing the metallic characteristics by improving the partial pressure of oxygen, and choosing right kind of ion source, the higher LIDT HfO_2 films can be prepared by ion-assisted reaction.

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