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Experiment and prediction on thermal conductivity of Al_2O_3/ZnO nano thin film interface structure

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Abstract. We predict that there is a critical value of Al_2O_3/ZnO nano thin interface thickness based on two assumptions according to an interesting phenomenon, which the thermal conductivity (TC) trend of Al_2O_3/ZnO nano thin interface is consistent with that of relevant single nano thin interface when the nano thin interface thickness is >300 nm; however, TC of Al_2O_3/ZnO nano thin interface is higher than that of relevant single nano thin interface when the thin films thickness is <10 nm. This prediction may build a basis for the understanding of interface between two different oxide materials. It implies an idea for new generation of semiconductor devices manufacturing.

Keywords. Al_2O_3/ZnO nano thin film; thermal conductivity (TC); nanoscale; interface structure.

1. Introduction

With the rapid development of micro/nano electronic technology and micro electromechanical systems, higher heat transfer requirements in micro/nano scale are put forward. Interface structure is one of the most important parts in micro/nano devices (Li and Zhang 2010). The thermal and optical properties of nano thin films have significant effect on the reliability and lifetime of micro/nano devices (Chiritescu *et al* 2007). The thermal properties are very different between nano thin films and bulk materials because of the size and boundary effects. TC (thermal conductivity) and optical properties of nano materials were investigated (Chiritescu *et al* 2007; Keppler *et al* 2008). TC of the Si nanowires was systemically studied under the different conditions of temperature (20–320 K) and diameters (22, 37, 56 and 115 nm). It indicated that the phonon scattering has influence on the thermal conductivity of Si nanowires (Li *et al* 2003). Liu *et al* (2011) investigated the TCs (thermal conductivities) of pure and Zn-doped $LiCu_2O_2$ single crystal; the results showed that the change of TC of $LiCu_2O_2$ single crystals was related with phonon scattering. Costescu *et al* (2004) investigated thermal properties of W/Al_2O_3 compound films under different thicknesses. It indicated that the compound film has ultra-low TC. Murshed *et al* (2008) and Glässl *et al* (2011) investigated TC of nanofluids. Lukes *et al* (2000) developed a study on TC of Ar solid thin films by using molecular dynamics. TC of Ar solid thin films increased with the increase of film thickness, which was consistent with the experimentally and theoretically predicted data. Abramson *et al* (2002) studied the interface and strain effects on TC of heterostructures using molecular dynamic method. It indicated that TC of thin films consisting of different materials is less than a half of the average TC. Yang and Liao (2008), (Yang *et al* 2011, 2012) and Liao *et al* (2010) investigated the heat transfer properties of interface structures based on multiscale model method. The interface thermal resistance and defects have significant effect on the thermal performance of interfacial structure. Choi *et al* (2006) tested TC of aluminum nitride and SiC thin films by using 3ω method. It indicated that TC of thin film was less than that of the corresponding bulk materials. At the same time, TC of thin film decreased with a decrease of thin film thickness.

The thermal performances of Al_2O_3/ZnO nano thin interfaces are studied in this paper. Al_2O_3 (Olsson *et al* 1999) and ZnO (Jin *et al* 2000; Fan *et al* 2006) materials are widely applied in microelectronics field because of their excellent optical and mechanical properties and thermostability (Xu *et al* 2010). Al_2O_3/ZnO samples are prepared by magnetic sputtering. The effects of the nanoscale and interface are examined experimentally and evaluated based on the experimental data.

2. Experimental

2.1 *Samples preparation*

Al_2O_3/ZnO nano thin films samples are deposited on silicon substrate by using radio frequency magnetron sputter-

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ing method at room temperature. The nano thin film thickness are 300, 500 and 800 nm, respectively. The parameters of experimental materials and samples of Al_2O_3/ZnO nano thin interface are shown in tables 1 and 2 as follows.

2.2 *Results*

TCs of Al_2O_3/ZnO thin film samples are measured by using transient thermo-reflectance technique (Bai *et al* 2008, 2009) and transmission-line theory (Hui and Tan 1994; Chen and Hui 1999). So, the heat conduction problem can be simplified to one-dimensional problem (Zhao *et al* 2004). The test system is shown in figure 1. The normalized temperature profiles of these samples, which are obtained by using transient and thermo-reflectance technique are shown in figure 2. TCs of Al_2O_3/ZnO thin film samples are obtained based on the test system, the results are shown in table 3.

The temperature distribution model of TC under frequency domain can be described as following:

$$T(s)=\frac{Q(s)}{e_1\sqrt{s}}\frac{\left[\begin{array}{l}\cosh\eta_1\sqrt{s}+e_{4,1}\sinh\eta_1\sqrt{s}\\+r_{\text{th}}e_4\sqrt{s}\cosh\eta_1\sqrt{s}\end{array}\right]}{\left[\begin{array}{l}\sinh\eta_1\sqrt{s}+e_{4,1}\cosh\eta_1\sqrt{s}\\+r_{\text{th}}e_4\sqrt{s}\sinh\eta_1\sqrt{s}\end{array}\right]},\tag{1}$$

where $Q(s) = 1$, which is the heat source. The temperature distribution model under time domain can be obtained by using the Laplace inverse transforms with the following function (Cheng *et al* 1994):

$$f(t)=\frac{\ln 2}{t}\sum_{n=1}^{N}C_n F\left(\frac{n\ln 2}{t}\right),\tag{2}$$

where

$$C_{\text{n}}=(-1)^{n+N/2}$$

$$\times\sum_{k=(n+1)/2}^{\min(n,N/2)}\frac{k^{N/2}(2k)!}{(N/2-k)!k!(k-1)!(n-k)!(2k-n)!};$$

$N = 8$, $k = [(n+1)/2]$.

This temperature distribution model is applied to match the test data shown in figure 3. It indicates that the fitting

Table 1. Parameters of experimental materials.

Materials	Purity	Size
α-Al_2O_3	99·99%	ϕ 50 mm × 3 mm
ZnO	99·99%	ϕ 50 mm × 3 mm
Si	*N*-type, single-sided polishing	0·5 mm

Table 2. Parameters of samples of Al_2O_3/ZnO nano thin interface.

Samples		Thickness (nm)		Nos.
Al_2O_3/ZnO	Al_2O_3	150	300	1
	ZnO	150		
Al_2O_3/ZnO	Al_2O_3	250	500	2
	ZnO	250		
Al_2O_3/ZnO	Al_2O_3	400	800	3
	ZnO	400		

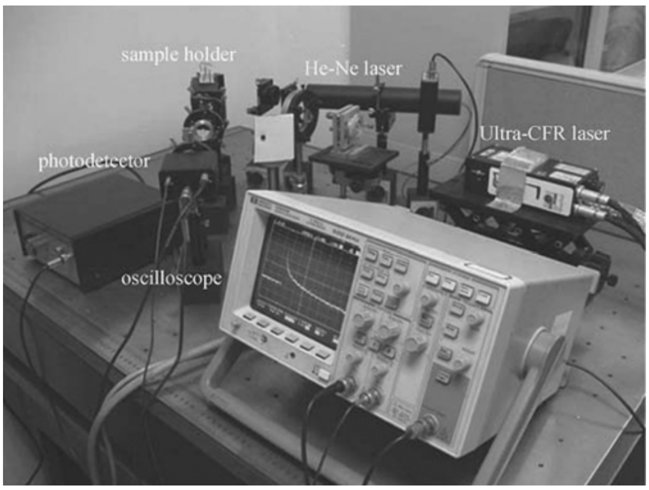

Figure 1. Principle of measurement and test system.

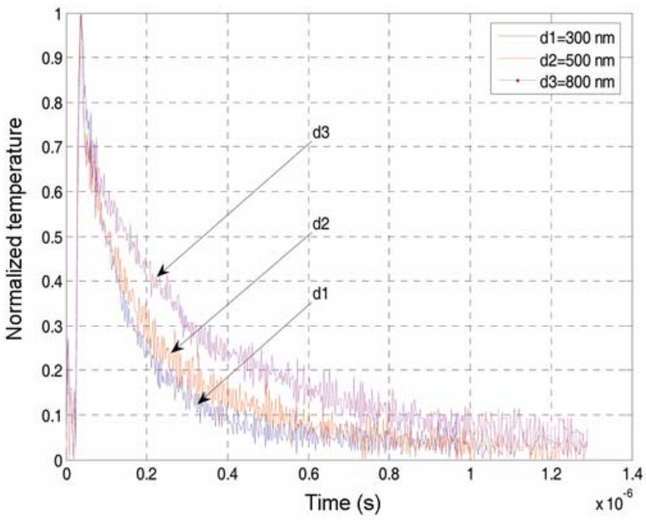

Figure 2. Transient surface temperature excursion profile of samples based on transmission-line theory and transient thermo-reflectance technique, *d* is thickness of Al_2O_3/ZnO thin film.

Table 3. TCs of Al_2O_3/ZnO thin film samples.

Thickness	300 nm	500 nm	800 nm
Thermal conductivity, *k* (W/m·K)	2·42	2·98	3·10

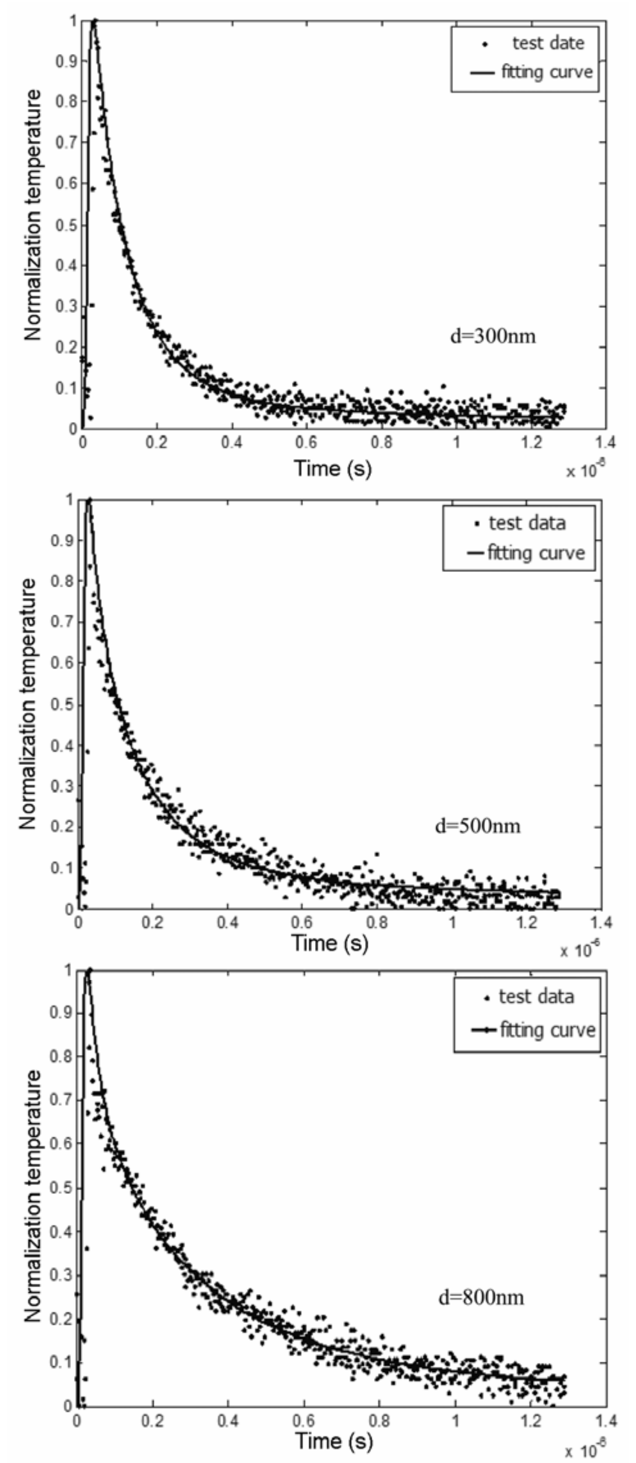

Figure 3. Fitting curves of temperature distribution model and test data under different film thicknesses.

model is matching with the test data. The decreasing rate of temperature is seen to slow down along with the nano thin interface thickness increase. The experimental TC of Al_2O_3/ZnO can be obtained by multi-points test. Meanwhile, the simulated method (Yang *et al* 2012) are used to calculate the numerical TC of Al_2O_3/ZnO, the final TCs are shown in table 3.

Table 4. Experimental and simulated data.

Thickness (nm)	TC (W/m·K)
Simulated TC of Al_2O_3/ZnO thin films	
2·34	1·26
2·86	1·44
3·30	1·53
4·60	1·67
5·20	1·79
Experimental TC of single Al_2O_3 thin films (Liu *et al* 2011)	
140	1·00
300	3·15
330	3·32
1000	3·47
Experimental TC of Al_2O_3/ZnO thin films	
300	2·42
500	2·98
800	3·10
Experimental TC of single ZnO thin films (Yang and Liao 2008)	
80	1·40
140	3·80
213	5·19
276	6·51

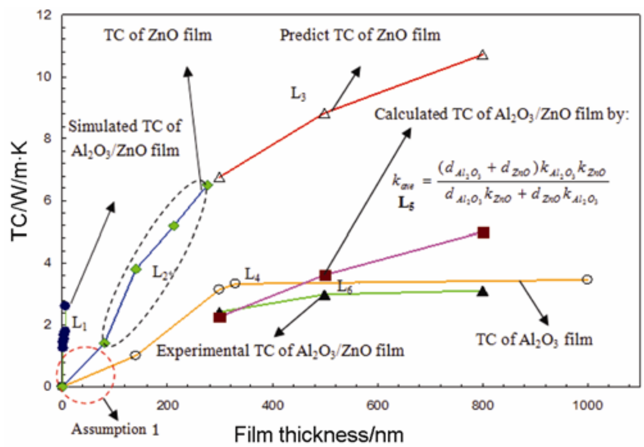

Figure 4. TC of Al_2O_3/ZnO nano thin interface, Al_2O_3 and ZnO nano thin films. L_1: curve of simulated TC of Al_2O_3/ZnO nano thin interface; L_2: TC of ZnO nano thin films (Yang and Liao 2008); L_3: predicted TC of single ZnO nano thin films based on experimental data (Yang and Liao 2008); L_4: TC of Al_2O_3 nano thin films; L_5: calculated TC of Al_2O_3/ZnO nano thin interface by using function (3); L_6: experimental TC of Al_2O_3/ZnO nano thin interface.

3. Assumptions and discussion

3.1 *Assumptions*

In order to contrast the simulated data with experimental data, two assumptions are put forward as follows: (a) When the thickness of thin films tend to zero, TC of thin films is tending to zero; (b) TC of thin films is similar with that of relevant bulk materials when the thickness of thin films is > 1000 nm. The experimental and simulated data used in this paper are shown in table 4.

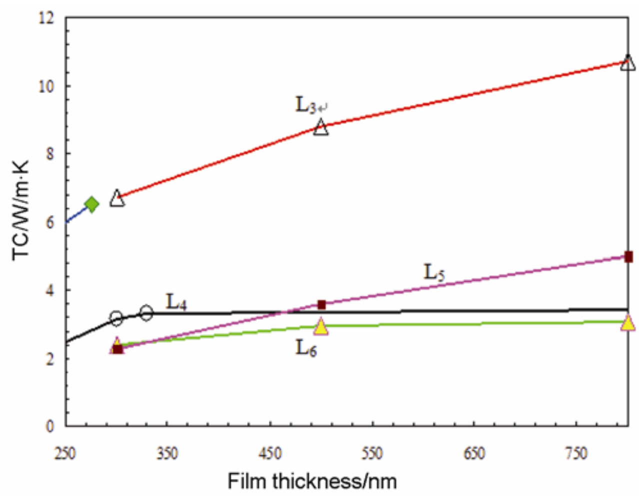

Figure 5. TC change of Al_2O_3/ZnO nano thin interface (film thickness > 300 nm). L_3: predicted TC of single ZnO nano thin films; L_4: experimental TC of Al_2O_3/ZnO film; L_5: curve of calculated TC of Al_2O_3/ZnO nano thin interface; L_6: curve of experimental TC of Al_2O_3/ZnO nano thin interface.

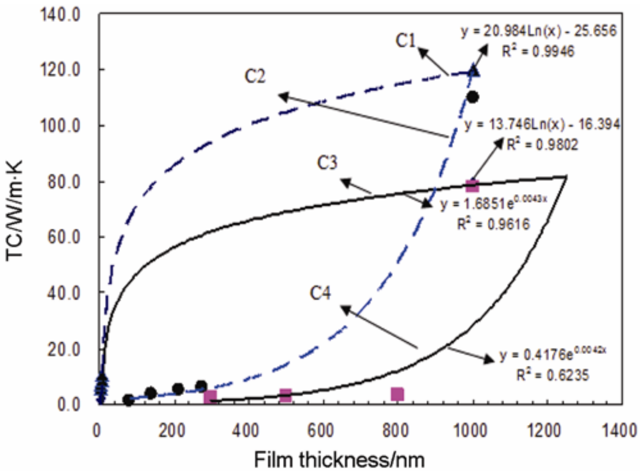

Figure 6. Fitting TC curves of Al_2O_3/ZnO nano thin interface compared with single ZnO nano thin interface. C_1: fitting curve of simulated TC of ZnO nano thin film; C_2: fitting curve of experimental TC of ZnO nano thin film; C_3: fitting curve of simulated TC of Al_2O_3/ZnO nano thin film; C_4: fitting curve of experimental TC of Al_2O_3/ZnO nano thin film.

3.2 *Discussions*

The average TC (k_{ave}) is introduced to verify the experimental and simulated results in this paper, which can be calculated by the following function (Abramson *et al* 2002):

$$k_{ave} = \frac{(d_{Al_2O_3} + d_{ZnO}) k_{Al_2O_3} k_{ZnO}}{d_{Al_2O_3} k_{ZnO} + d_{ZnO} k_{Al_2O_3}}, \quad (3)$$

where $d_{Al_2O_3}$ is the thickness of Al_2O_3 nano thin film. d_{ZnO} is the thickness of ZnO nano thin film. $k_{Al_2O_3}$ is the TC of Al_2O_3 nano thin film. k_{ZnO} is the TC of ZnO nano thin film. The average TC of Al_2O_3/ZnO nano thin interface can be calculated by (3). TC curves of Al_2O_3/ZnO nano thin films, Al_2O_3 nano thin films and ZnO thin films are shown in figure 4. In figure 4, L_1 is the curve of simulated TC of Al_2O_3/ZnO nano thin interface; L_2 is TC of ZnO nano thin films (Yang and Liao 2008); L_3 is the prediction TC of single ZnO nano thin films based on the experimental data (Yang and Liao 2008); L_4 is the TC of Al_2O_3 nano thin films; L_5 is the calculated TC of Al_2O_3/ZnO nano thin interface by using (3); L_6 is the experimental TC of Al_2O_3/ZnO nano thin interface.

From figure 4, the changing trend of TC of Al_2O_3/ZnO nano thin interface is consistent with that of the single ZnO and Al_2O_3 nano thin films (Bai *et al* 2008; Huang *et al* 2011). Meanwhile, it is similar with the calculated average TC (k_{ave}) while the nano thin films thickness is > 300 nm shown in figure 5. Three feasible reasons can be explained this as follows: (a) When the thickness of nano thin films is > 300 nm, the size effect is not significant; (b) Although there is an error between experimental TC and calculated average TC of Al_2O_3/ZnO nano thin interface because of samples purity, test error, fitting

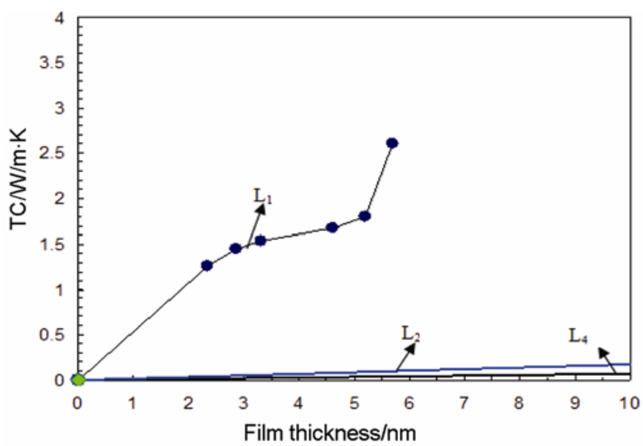

Figure 7. TC change of Al_2O_3/ZnO nano thin interface, Al_2O_3 and ZnO nano thin interface (film thickness < 10 nm). L_1: curve of simulated TC of Al_2O_3/ZnO nano thin interface; L_2: curve of TC of ZnO thin films; L_4: curve of TC of Al_2O_3 nano thin films.

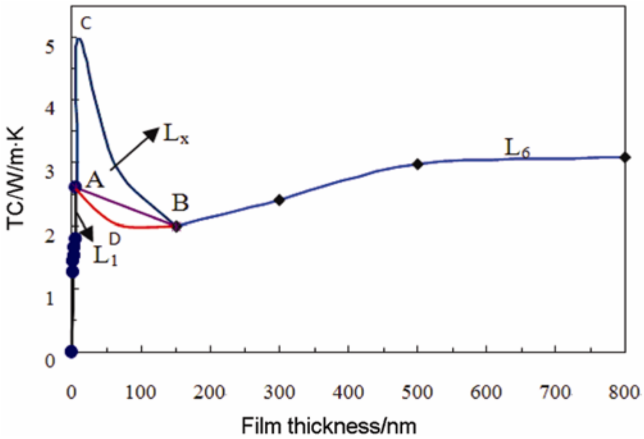

Figure 8. Predicting TC curves of Al_2O_3/ZnO thin films. L_1: curve of simulated TC of Al_2O_3/ZnO nano thin interface; L_6: curve of experimental TC of Al_2O_3/ZnO nano thin interface; L_x: artificial lines (ACB, AB, ADB) between A and B.

error, the change trend of TC of Al_2O_3/ZnO nano thin interface is similar with that of the single nano thin film and the average TC; (c) TC of Al_2O_3/ZnO nano thin interface is less than that of the single ZnO nano thin film due to the nano thin interface doped Al_2O_3 materials of low TC shown in figure 6. It indicates that the trend of TC of Al_2O_3/ZnO nano thin film agrees with that of single ZnO nano thin film.

When the thickness of nano thin interface is < 10 nm, TC of Al_2O_3/ZnO nano thin interface is higher than that of each single thin film of Al_2O_3 or ZnO. This is abnormal performance compared with that of the nano thin interface while the thickness is > 300 nm. This is an interesting phenomenon shown in figure 7.

3.3 *Prediction*

The fitting curves of experimental and simulated data based on the least square method are shown in figure 8. There are three possible changes between points A and B as follows: (a) ACB. In this case, based on the fitting model of simulation data, the changing trend of L_1 is increasing from A position until a certain value (C position), and then it is decreasing to B position. (b) AB. the changing trend of L_1 from points A to B is direct. (c) ADB. Based on the fitting model of experimental data, the changing trend of L_1 is decreasing from the position A until a certain value (D position) and then it is increasing to the position B.

No matter in any case, there must be a critical value of thickness of Al_2O_3/ZnO nano thin interface based on the fitting model of the experimental and simulation data verified by (Bai *et al* 2008; Huang *et al* 2011). There is also a critical TC of Al_2O_3/ZnO nano thin interface at this critical thickness. Four possible reasons are put forward according to this phenomenon as follows: (a) The size effect of interface thermal resistance of Al_2O_3/ZnO nano thin interface is obvious at ultra-thin film thickness; (b) Although the phonon scattering is increasing (Flik *et al* 1992; Zeng and Chen 2001; Kim *et al* 2011), the phonons are easy to cross the thin films interface structure in this scale when the thin films thickness is less than the phonon mean free path. The effect of phonon through the interface structure is more important than the phonon scattering. So, the increase of heat transfer in unit time TC is increasing; (c) In this nanoscale, the atom numbers of Al_2O_3/ZnO nano thin interface are relatively increasing compared with the single nano thin film. Hence, hot carriers at the moment are relatively increasing, which cause the increase of TC; (d) The lattice of Al_2O_3/ZnO nano thin film is mismatching under the ultra-thickness (Kim *et al* 2011), which could cause an increase of TC of the Al_2O_3/ZnO nano thin interface.

4. Conclusions

A critical thickness value of Al_2O_3/ZnO nano thin interface is predicted based on the fitting models of experimental and simulation data. Four reasons, interface resistance, the ability of phonon through the interface structure, relative thermal carriers numbers and lattice mismatching are put forward to explain this phenomenon. It means that the TC of Al_2O_3/ZnO nano thin interface is relatively highest at this critical thickness value. It will be one of the most important parameters in microelectronic manufacturing.

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