THIN FILMS THERMAL CONDUCTIVITY VERSUS
SUBSTRATE SURFACE ROUGHNESS

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Abstract

The thermal conductivity and the interface thermal contact resistance of four different thickness CuO films, obtained by PVD on a tungsten carbide substrate were investigated. The periodic photothermal experiment based on infrared measurement was developed. According to the knowledge of the substrate thermophysical properties the gain between the thermal perturbation and the response was used as the experimental data. The results show the dependence of the thermal conductivity on the film thickness. The scanning electron microscopy observations present strong anisotropy and columnar structure of the deposits. Moreover, the chemical composition of films was tested. The presence of cobalt diffused from the substrate was revealed, and similar composition for each film was found.

Keywords: Thin Film, Thermal Conductivity, Radiative Photothermal Experiment, Thermal Contact Resistance
Introduction

The thermal conductivity estimation of thin solid films of sub micrometric thickness on the form of a coating is well treated in the literature. There exist different experimental approaches all based on the front face technique. It consists in measuring the temperature at the location of the thermal disturbance that is a heat flux (see figure 1). The 3-ω technique is perhaps the simplest experimental method [1]. It consists in depositing an Au layer on the deposit whose thermal conductivity is unknown. A periodic current passes through the Au layer that induced Joule effect and heat transfer in the deposit and the substrate. Given that the Au electrical resistance depends on the temperature, the Au layer is used as the heater and as the thermal sensor. Indeed, this resistance can be measured by inserting the Au layer in a Wheatstone bridge. A comparable method is used in [2] but its configuration differs from the previous one by focusing a pico-second laser beam on the Au layer that is covered by a constant current. This technique permits exploring highest frequencies and lead to estimate the thermal conductivity as well of the thermal diffusivity of the deposit. In the experimental configuration developed by [3], the surface temperature measurement is reached using the thermo-reflectance technique. The Au layer is well appropriate for such an experiment given it offers a very interesting thermo reflection coefficient. As previously, this experiment can also lead estimating the thermal diffusivity. Furthermore, surface temperature can be obtained using the photothermal infrared radiometry technique. Obviously in that case, the Au layer disappears and heating is obtained by focusing a laser beam on the material.

Two classical heat flux waveforms are used in such thermal characterization experiments: the periodic one and the impulse one. As presented in [4] and [5], each approach has its own advantages and drawbacks. It must be also cited the use of an intermediate approach based on the use of a random heat flux [6]. In practice, the impulse technique requires only one
experiment as the periodic method requires an experiment for each explored frequency. Nevertheless, this last method leads to the best results in terms of reliability and estimated parameters confidence domains. A sensitivity study is performed in order to precisely define the limits of the frequency domain according to the parameters that will be estimated (thermal conductivity and diffusivity).

Whatever the choice of the experimental technique and heat flux disturbance, the result is the same: the thermal conductivity of the thin layer strongly varies according to its thickness. This conductivity is all the more small as the thickness is itself small. Cahill and Lee [1] and showed this phenomenon on deposits of SiO₂, TiO₂, Al₂O₃, HfO₂ and MgO. Orain et al. [2] find a comparable tendency for a ZrO₂ film. Bhusari et al. [7] obtain also this decrease for an aluminium deposit, of which the thickness varies from 0.5 to 2.5 μm, on a silicon and copper substrate. On the contrary, Langer et al. [8] show that the thermal conductivity of a nickel film on a quartz substrate does not vary according to the thickness.

Several reasons are called upon to explain this variation of conductivity according to the thickness. First of all, when metal layer thickness is lower than 10-100 nm, the mean free path of the carriers (phonons and electrons) does not make possible the definition of thermal conductivity within the meaning of the Fourier law. In that case, it is necessary to carry out a model of molecular dynamics scale, and thermal conductivity is that of the lattice [9][10]. For larger thicknesses, the variation of conductivity comes from the considerable influence of the deposit-substrate interface. The imperfect contact between materials is modelled in the form of a thermal contact resistance. This resistance is related to many parameters but in fact the parameters (temperature, pressure) related to the process of deposition (PVD, CVD) have the most significant contribution. It was shown for a long time the influence of the deposition temperature on the arrangement of the constitutive particles of the layer and on its density [11]. In the same way, particles column-like arrangement is found at critical temperatures. In
that case, it would be judicious to consider an anisotropic thermal conductivity, in any event incomparable with that of the bulk material. Lastly, when the substrate is not chemically any more neutral with respect to the deposit, one must take into account species diffusion phenomenon with the deposit-substrate interface.

The objective of this work is to show the relation between the thermal conductivity of the deposit, its structure, its chemical composition and the surface roughness of the substrate. For that, our step is built around the characterization of a CuO deposit which one varied the thickness. The coating is deposited by PVD on a WC-Co9% substrate. It has an average roughness (~300 nm) and cobalt diffuses rather easily in the deposit at the time of the deposition process.

The total surface emissivity of the CuO is close to 0.8 in the 0-100°C temperature range [12]. Such a feature improves incident radiation absorption and infrared radiation emissivity. Thereby, the experimental method for the thermal conductivity estimation is based on the periodic photothermal infrared radiometry.

**Modulated photothermal IR radiometry experiment**

The schematic view of the measurement arrangement is shown in figure 2. The system is composed of two parts: the sample excitation and the thermal response measurement. The thermal excitation is generated on the deposit front face from a laser diode of 808 nm wavelength and 5 W maximal power. The laser is modulated directly from its driver using sinusoidal signal from the frequency function generator. Such external modulation range is up to 20 kHz. Focused on the mirror by a convex lens the laser beam is redirected perpendicularly to the face of the studied sample. The laser beam has a Gaussian profile as specified from the constructor. A very fast photodiode is used to measure the reference signal in order to avoid the phase lag due to the laser diode driver.
The thermal response is measured by HgCdTe based photovoltaic infrared detector, cooled down to liquid nitrogen temperature (77K). The measurement wavelength range is 5-13 µm (see figure 3). The elliptical mirror coated with high reflective rhodium (reflectivity 98% in the infrared detector wavelength band – see figure 3) is used to collect the emitted infrared radiation and focus it on the infrared detector. In fact, the heated surface of the sample is located at interior focal point of the mirror and the sensitive element of the detector at the exterior one, what results in a conjugate focuses system. The incident laser beam is centred and aligned on the focal axis of the mirror. The detector wavelength band is largely greater than that of the laser, what gives the advantage that the measurement is not disturbed by the excitation source. The zone viewed from the detector is the image of the infrared sensitive element on the sample; it approximately corresponds to circle of 1 mm diameter. As represented in figure 4, the HgCdTe detectivity is null when the frequency of the measured signal is less than 10 Hz and it is constant from 10 Hz until 10 KHz. Thereby, the detector is not sensitive to the constant component of the measured signal. The signal from the detector is amplified with an adapted preamplifier, and then is visualized on a digital oscilloscope with the photodiode signal. A lock-in amplifier is used to measure the gain and the phase lag between the reference and the detector signals as the frequency function. The signal from the infrared detector is exploitable up to 2 kHz. As demonstrated after in the sensitivity study, this value is sufficient in order to estimate the thermal conductivity of the deposit. The noise variance for the IR detector and the photodiode in the 10-2000 Hz frequency range had been measured and reported in table 1. A constant variance is found for the two sensors over this frequency interval. The literature [12] gives a CuO surface emissivity close to 0.8 but it varies according to the deposit thickness, to the deposition process parameters and the surface roughness. Its exact determination involves specific experiment. On the other hand, the
incident heat flux can not be easily measured. Thus, the chosen available measurable quantities are the voltage at the HgCdTe sensor and at the photodiode.

**Heat transfer modelling**

According to the experimental cylindrical geometry represented in figure 1, the temperature $T(r, z, t)$ at each point of the medium is given from the governing equations of heat transfer in the deposit and the substrate:

$$\begin{cases}
\left( \rho C_p \right)_d \frac{\partial T}{\partial t} = \frac{\lambda_d}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + \lambda_d \frac{\partial^2 T}{\partial z^2}, & 0 < z < e_d \\
\frac{1}{a_s} \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial T}{\partial r} \right) + \frac{\partial^2 T}{\partial z^2}, & e_d < z < e_d + e_s
\end{cases}, \quad 0 < r < R, t > 0 \tag{1}
$$

$\lambda_d$ and $\lambda_{ds}$ are the thermal conductivity of the deposit in the radial and longitudinal directions respectively, $\left( \rho C_p \right)_d$ is the volumic heat of the deposit and $a_s$ is the thermal diffusivity of the substrate, that is assumed to be homogenous given to the measurement area.

The boundary condition at the surface of the deposit is:

$$-\lambda_d \frac{\partial T}{\partial z} = \begin{cases}
\phi_0(r, t), & 0 < r \leq r_0, z = 0 \\
hT, & r_0 < r < R, z = 0
\end{cases}, \quad t > 0 \tag{2}
$$

$h$ is the heat exchange coefficient and $\phi_0$ is the heat flux density from the laser. The cylindrical symmetry and insulated condition at the periphery lead to the following relation:

$$\frac{\partial T}{\partial r} = 0, \quad r = 0, r = R, 0 < z < e_d + e_s, t > 0 \tag{3}
$$

Heat exchange at the interface between the substrate and the ambient is written as:

$$-\lambda_s \frac{\partial T}{\partial z} = hT, \quad 0 < r < R, z = e_d + e_s, t > 0 \tag{4}
$$

The thermal contact resistance $R_c$ at the deposit-substrate interface is defined as:

$$T_d - T_s = R_c \phi, \quad 0 < r < R, z = e_d, t > 0 \tag{5}$$
Finally, the initial condition is:

\[ T = 0, \quad 0 \leq r \leq R, 0 \leq z \leq e_d + e_s \]  \quad (6)

Applying the Fourier transform, versus the time variable, on the temperature and the heat flux density lead to:

\[
\theta(r, z, j\omega) = \int_0^\infty T(r, z, t)e^{-j\omega t} \, dt, \quad \psi(r, j\omega) = \int_0^\infty \varphi(r, t)e^{-j\omega t} \, dt
\]  \quad (7)

Thereby, relations (1) to (6) become:

\[
\begin{align*}
\left( \rho C_p \right)_d s\theta & = \frac{\lambda_d}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \theta}{\partial r} \right) + \lambda_{de} \frac{\partial^2 \theta}{\partial z^2}, \quad 0 < z < e_d, \\
\frac{s}{a_s} \theta & = \frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial \theta}{\partial r} \right) + \frac{\partial^2 \theta}{\partial z^2}, \quad e_d < z < e_d + e_s
\end{align*}
\]  \quad (8)

\[
-\lambda_d \frac{\partial \theta}{\partial z} = \begin{cases} \omega_0, & 0 < r \leq r_0, z = 0 \\ h\theta, & r_0 < r < R, z = 0 \end{cases}
\]  \quad (9)

\[
\frac{\partial \theta}{\partial r} = 0, \quad r = 0, r = R, 0 < z < e_d + e_s
\]  \quad (10)

\[
-\lambda_s \frac{\partial \theta}{\partial z} = h\theta, \quad 0 < r < R, z = e_d + e_s
\]  \quad (11)

\[
\theta_d - \theta_s = R\psi, \quad 0 < r < R, z = e_d
\]  \quad (12)

According to the cylindrical symmetry, one can apply the Hankel transform on the temperature and the heat flux density, versus the radial coordinate, as:

\[
\tilde{\theta}(\alpha, z, s) = \int_0^\infty \theta(r, z, s) r J_0(\alpha r) \, dr, \quad \tilde{\psi}(\alpha, s) = \int_0^\infty \psi(r, s) r J_0(\alpha r) \, dr
\]  \quad (13)

Thus, relations (8) to (12) become:

\[
\begin{align*}
\left( \rho C_p \right)_d s\tilde{\theta} & = -\alpha^2 \lambda_d \tilde{\theta} + \lambda_{de} \frac{\partial^2 \tilde{\theta}}{\partial z^2}, \quad 0 < z < e_d, \\
\frac{s}{a_s} \tilde{\theta} & = -\alpha^2 \tilde{\theta} + \frac{\partial^2 \tilde{\theta}}{\partial z^2}, \quad e_d < z < e_d + e_s
\end{align*}
\]  \quad (14)
\[-\lambda_d \frac{\partial \bar{\psi}_o (\alpha,t)}{\partial z} = \begin{cases} \bar{\psi}_o (\alpha,t), & z = 0 \\ h \bar{\theta}, & z = 0 \end{cases} \tag{15}\]

\[J_i (\alpha R) = 0, \quad 0 < z < e_d + e_s \tag{16}\]

\[-\lambda_s \frac{\partial \bar{\psi}}{\partial z} = h \bar{\theta}, \quad z = e_d + e_s \tag{17}\]

\[\bar{\theta}_d - \bar{\theta}_s = R \bar{\psi}, \quad z = e_d \tag{18}\]

Relation (16) is a transcendental equation whose solutions are approximately:

\[\alpha_n R \approx \pi \left( n + \frac{1}{4} \right) - \frac{3}{8 \pi \left( n + \frac{1}{4} \right)}, \quad \alpha_0 = 0 \tag{19}\]

It has been shown in the previous section that the laser beam has a Gaussian profile:

\[\varphi_o (r,t) = e^{-\left( \frac{r}{a_0} \right)^2} f(t) \tag{20}\]

Applying respectively the Fourier and Hankel transforms on relation (20) lead to:

\[\bar{\psi}_o (\alpha, j\omega) = e^{-\frac{a^2r^2}{4}} F (j\omega) \tag{21}\]

The solution of equation (14) is on the form:

\[\bar{\theta}_i (\alpha, z,s) = A_1 e^{h z} + A_2 e^{-h z}, \quad i = d, s \tag{22}\]

Where \(A_1\) and \(A_2\) would be determined from boundary conditions (15) and (18), and:

\[k_s = \sqrt{s + \alpha_0^2} \quad \text{and} \quad k_d = \sqrt{\frac{(\rho C_p)_d s}{\lambda_d} + \frac{\lambda_0}{\lambda_d}} \alpha_0^2 \tag{23}\]

Taking \(z = 0\) in relation (22), one obtain the expression of the transfer function that expresses the transformed temperature at the heated surface according to the transformed heat flux as:

\[\frac{\bar{\theta}_0}{\bar{\psi}_o} = \frac{\beta}{1 - h \beta} \tag{24}\]

With:
\[ \beta = \frac{A + hB}{C + hD} \]  

And:

\[
\begin{align*}
A &= \cosh(k_d e_d) \cosh(k_s e_s) + \left( \cosh(k_d e_d) R_c + \frac{\sinh(k_d e_d)}{\lambda_d k_d} \right) \lambda_s k_s \sinh(k_s e_s) \\
B &= \frac{\sinh(k_s e_s)}{\lambda_s k_s} \cosh(k_d e_d) + \left( \cosh(k_d e_d) R_c + \frac{\sinh(k_d e_d)}{\lambda_d k_d} \right) \cosh(k_s e_s) \\
C &= \lambda_d k_d \sinh(k_d e_d) \cosh(k_s e_s) + \left( \lambda_d k_d \sinh(k_d e_d) R_c + \cosh(k_d e_d) \right) \lambda_s k_s \sinh(k_s e_s) \\
D &= \left( \lambda_d k_d \sinh(k_d e_d) R_c + \cosh(k_d e_d) \right) \cosh(k_s e_s) + \frac{\lambda_d k_d}{\lambda_s k_s} \sinh(k_d e_d) \sinh(k_s e_s)
\end{align*}
\]

Using the thermoelectric analogy gives more insight on the heat transfer transient behaviour as represented on the top of figure 5. \( Z_d \) and \( Z_s \) respectively denote the deposit and substrate impedances.

**Low frequency behaviour and sensitivity study**

At low heat flux frequencies, \( f \ll a_d / e_d^2 \), the deposit is viewed as a thermal resistance and the transfer function can be largely simplified given that parameters \( A, B, C \) and \( D \) of relation (26) become:

\[
\begin{align*}
A &= \cosh(k_s e_s) + R_d \lambda_s k_s \sinh(k_s e_s) \\
B &= \frac{\sinh(k_s e_s)}{\lambda_s k_s} + R_d \cosh(k_s e_s) \\
C &= \lambda_s k_s \sinh(k_s e_s) \\
D &= \cosh(k_s e_s)
\end{align*}
\]

In this relation the thermal resistance \( R_d \) is expressed according to the thermal conductivity in the longitudinal direction and the thickness of the deposit and the thermal contact \( R_c \) as:

\[ R_d = \frac{e_d}{\lambda_d} = \frac{e_d}{\lambda_d} + R_c \]  

The equivalent electrical scheme is represented at the bottom of figure 5.
Applying the inverse Hankel transform on relation (24) lead to:

\[
\theta_0(r, j\omega) = \left( \sum_{n=1}^{\infty} \frac{2 J_n(\alpha_n r)}{R^2 J_0(\alpha_n R)} e^{-\frac{\alpha_n^2 r^2}{4}} e^{-\frac{\beta}{1-h\beta}} \right) F(j\omega)
\]

The average temperature on the measurement area is:

\[
\langle \theta_0(j\omega) \rangle = \frac{1}{r_m^2} \int_0^{r_m} \theta_0(r, j\omega) r \, dr
\]

With respect to relation (29), one obtains an analytical expression of the average temperature on the measurement area as:

\[
\langle \theta_0(j\omega) \rangle = \begin{cases} 
\left( \sum_{n=1}^{\infty} \left( \frac{2}{r_m R} \right)^2 e^{-\frac{\alpha_n^2 r_m^2}{4}} \frac{\beta}{1-h\beta} \right) F(j\omega), & n = 0 \\
\left( \sum_{n=1}^{\infty} \frac{4 J_n(\alpha_n r_m)}{r_m \alpha_n R^2 J_0(\alpha_n R)} e^{-\frac{\alpha_n^2 r_m^2}{4}} e^{-\frac{\beta}{1-h\beta}} \right) F(j\omega), & n > 0
\end{cases}
\]

That can be written on the transfer functional form:

\[
\langle \theta_0(j\omega) \rangle = Z(j\omega) F(j\omega)
\]

We use the classical definition of the gain of the transfer function as:

\[
G(j\omega) = 20 \log_{10} \| Z(j\omega) \|
\]

As said in the previous section, the heat flux and the temperature can not be estimated accurately. The measurement of the photodiode and the infrared detector voltage leads to the gain proportional to that between the heat flux and the temperature. The sensitivity study for the gain evolution versus the frequency and the thermal properties permits to overcome this drawback. In fact the low frequency behaviour of the deposit-substrate system for a large thermal conductivity range is identical as the uncoated substrate. Such behaviour permits to scale the measured gain to the real one. The known thermal parameters (see table 2) are used to plot the gain for different film conductivity values over the frequency interval 1-2000 Hz, as represented in figure 6. The gain variation versus frequency for \(10 < f < 2000\) is small but
sufficient with regards to the measurement error in order to estimate the deposit thermal conductivity.

**Results**

**Deposit morphology**

The Wc-Co9% substrate is 16 mm diameter and 1 mm thickness. Uniformity of the deposition surface roughness is reached from high pressure jet of calibrated Al₂O₃ particles (~3 µm diameter). Surface is then cleaned by exposure to the ultrasounds. The surface morphology has been measured using an optical interferometer device. Explored area is (80µm×60µm) and the measurement has been repeated on three different zones of the surface in order to verify the roughness uniformity. The 3D surface morphology is represented on figure 7. The calculated average roughness is: \( R_a = 300 \text{ nm} \).

The CuO is deposited using the PVD technique. Process parameters are: RF power density 1.7 W/cm², substrate temperature 25°C, deposition rate: 44.8 nm/min, total pressure: 0.5 Pa oxygen, partial pressure 0.03 Pa (6%), crystallisation state: amorphous. Four samples have been realized, considering different deposit thickness: 0.5 µm, 0.75 µm, 1 µm and 1.58 µm. As previously, the 3D surface morphology has been measured for each sample and results are presented in figure 8, roughness average values are reported in table 3. As one can see, the roughness does not significantly vary whatever the deposit thickness. This result shows that the deposit follows the substrate roughness. The film structure has been studied using Scanning Electron Microscopy (SEM) apparatus. As represented on figure 9, the result is a tapered columnar structure with the columns ending in domed tops as suggested in figure 1. This is coherent with respect to the value of the temperature deposition: \( T/T_m < 0.3 \) (where \( T_m \) is the CuO melting temperature). Whatever the deposit thickness, the average grain size is
approximately 1 µm and the deposit appears to be clearly anisotropic. Chemical composition of each deposit is also measured using SEM and results are reported in table 4. It concerns an average value in a 1µm thick layer from the deposit surface. It appears that the CuO composition is homogeneous whatever the deposit thickness. Nevertheless, a small quantity of Cobalt is observed even when the deposit thickness is greater than the explored depth. This means that cobalt has diffused in the deposit during the deposition process. This result is confirmed from the Auger experiment whose results are presented in figure 10.

**Deposit conductivity**

The periodic photothermal experiments were carried out on four CuO film samples. The explored frequency range is comprised between 10 Hz and 2 KHz, it was chosen according to the sensitivity analysis presented in the previous section. The thermophysical properties of the substrate were identified using the Hot Disk technique [13] for the thermal conductivity and the Differential Scanning Calorimetric technique for the specific heat. The \( \rho C_p \) product for the deposit was identified by the Differential Scanning Calorimetric for \( C_p \) and volumetric apparatus for \( \rho \) using the artificially obtained CuO crystals. These properties are reported in table 2 with their standard deviations. The heat exchange with the ambient characterized by \( h \) coefficient is: 5 W.m\(^{-2}\).K\(^{-1}\).

The deposit longitudinal thermal conductivity is identified by minimizing the quadratic gap between the theoretical and experimental values of the gain. The algorithm is a subspace trust region method and is based on the interior-reflective Newton method described in [15][14]. Each iteration involves the approximate solution of a large linear system using the method of preconditioned conjugate gradients (PCG). Figure 11 represents the comparison of the measured gain and the fitted one for each CuO film thickness.
The value of $R_d = e_d / \lambda_{di}$, where $\lambda_{di}$, have been identified for each thickness (table 5), are reported in figure 12. According to relation (28), it is found by taking $e_d = 0$ that $R_c = 4.20 * 10^{-7} \pm 1.77 * 10^{-4}$ m$^2$.K.W$^{-1}$ and thus the intrinsic value of the longitudinal thermal conductivity is $\lambda_{di} = 3.72 \pm 0.286$ W.m$^{-1}$.K$^{-1}$.

**Discussion**

The study permits identifying the longitudinal thermal conductivity a deposit on a substrate. Obviously, the substrate thermophysical parameters must be known precisely in order to obtain a reliable result. The investigation of the deposit-substrate interface thermal contact resistance may provide information about the quality of the contact. The layer of the deposit constitutes a thermal resistance that is considered as the sum of the intrinsic thermal resistance of the film and the thermal contact resistance. The intrinsic resistance concerns the thermal conductivity of the film material without defects. The thermal contact resistance include the defects in the transition layer and the interface deposit –substrate.

The described method was used to measure the thermal conductivity of a series of CuO films as the function of the thickness. The intrinsic thermal conductivity of CuO film was found lower than 4 W.m$^{-1}$.K$^{-1}$. The thermal conductivity of bulk CuO is of 17 W/K/m order in the literature. The obtained results reveal that the thermal conductivity of film decreases with decreasing film thickness. This difference are explained by the deposit structure. The optical interferometry, scanning electron microscopy and chemical composition analysis were performed in order to test the film morphology. The found anisotropic and columnar structure proves the existence of the thermal barriers between the grains that affects the lateral heat transfer which leads to thermal conductivity decrease. The Auger and SEM analysis revealed similar chemical composition for each film, therefore there is no influence from the chemical point of view. A special attention should be paid to the roughness of the substrate. The
roughness does not change after deposition process, what signify that the coating follows exactly the substrate form. In the thermal analysis, a plane surface of the film is considered. In reality, at the scale of µm, this surface is constituted of peaks and valleys covered by the film columns. The identified thermal conductivity of such deposit can be viewed as the average between the longitudinal and the transversal one, especially that the experimental technique leads to measurement on 1 mm diameter disk. The presented modeling of the heat transfer in the studied system distinguishes two-directional conductivity. The identification of plane conductivity will be extension of this work.

The morphology and chemical composition of thin film depend on the deposition process parameters [11]. Certainly, the thermal conductivities of films realized in different condition will be different. Moreover, the evolution of thermal conductivity according to thickness may also differ between series realized in different condition.

Conclusion

The periodic technique for the thermal conductivity measurement was presented. It is based on gain measurement from an infrared photothermal experiment. An optimum modulation frequency range was defined. The experimental constraints limit it to 2 kHz. The study shows similar low frequency behaviour for the uncoated substrate and the deposit-substrate system. This behaviour permits to scale the measured gain to the real one in the thermal diffusion process. The axis-symmetric model of heat transfer was used to fit the experimental data, and identify the longitudinal thermal conductivity of the film, considering the exact knowledge of the substrate parameters. The measurement on deposits with various thickness shows the thickness dependence of the thermal conductivity of CuO films on tungsten carbide substrates. It decreases linearly as the film thickness decreases from 1580 nm to 500 nm.
Considering the film as the thermal resistance has led determine the thermal contact resistance between the deposit and the substrate and the longitudinal intrinsic thermal conductivity of the film. As previously, the thermal resistance presents strong thickness dependence. In fact thin film deposited by PVD or CVD on a substrate largely differs in its structure from a solid material. A study concerning the films of the same thickness deposited at different temperatures and so on with different morphologies will be complement to this work.

References


Figure 1: Deposit thermal characterisation using the front face experiment. A transient heat flux density \( \varphi \) is applied on the deposit surface (over the disc of radius \( r_0 \)) and one measures the average temperature \( \langle T_0 \rangle \) on the heated area.
Figure 2: Schematic view of the experimental setup.
Figure 3: Spectral response of the infrared detector and the mirror reflectivity in function of the wavelength.
Figure 4: HgCdTe infrared detector responsivity and noise versus frequency.
Figure 5: Corresponding electrical analogy of the heat transfer model in the deposit-substrate system.
Figure 6: Sensitivity of gain versus $\lambda_{dz}$ on the $[10 – 2000]$ Hz frequency range.

\[ \lambda_s = 41 \text{ Wm}^{-1} \text{K}^{-1} \], \( \left( \rho C_p \right)_s = 2.938 \times 10^6 \text{ Jm}^{-3} \text{K}^{-1} \), \( e_s = 1 \text{ mm} \), \( \left( \rho C_p \right)_d = 2.898 \times 10^6 \text{ Jm}^{-3} \text{K}^{-1} \), \( e_d = 1 \mu\text{m} \), \( r_o = 1.5 \text{ mm} \), \( r_m = 0.5 \text{ mm} \), \( h = 5 \text{ Wm}^{-2} \text{K}^{-1} \).
Figure 7: Wc-CO surface morphology using optical interferometer.
Figure 8: Surface morphology using the optical interferometer.
Figure 9: Deposit structure from Scanning Electron Microscopy.
Figure 10: Auger experiment.
Figure 11: Comparison between the measured gain and fitting.
**Figure 12:** Evolution of the apparent thermal contact resistance with thickness.
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<td>500</td>
<td>0.062</td>
<td>6.10E-05</td>
</tr>
<tr>
<td>600</td>
<td>0.056</td>
<td>6.30E-05</td>
</tr>
<tr>
<td>700</td>
<td>0.053</td>
<td>6.20E-05</td>
</tr>
<tr>
<td>800</td>
<td>0.053</td>
<td>6.09E-05</td>
</tr>
<tr>
<td>900</td>
<td>0.052</td>
<td>6.26E-05</td>
</tr>
<tr>
<td>1000</td>
<td>0.049</td>
<td>6.01E-05</td>
</tr>
<tr>
<td>1200</td>
<td>0.047</td>
<td>6.21E-05</td>
</tr>
<tr>
<td>1400</td>
<td>0.046</td>
<td>6.35E-05</td>
</tr>
<tr>
<td>1600</td>
<td>0.045</td>
<td>6.19E-05</td>
</tr>
<tr>
<td>1800</td>
<td>0.041</td>
<td>6.65E-05</td>
</tr>
<tr>
<td>2000</td>
<td>0.04</td>
<td>6.26E-05</td>
</tr>
</tbody>
</table>

*Table 1:* The infrared detector and the photodiode noise variance for each explored frequency.
<table>
<thead>
<tr>
<th>Material</th>
<th>Thickness $e_s$ [mm]</th>
<th>Diameter $2R$ [mm]</th>
<th>Density $\rho$ [kg.m$^{-3}$]</th>
<th>Thermal conductivity $\lambda$ [W.m$^{-1}$.K$^{-1}$] (Hot Disc)</th>
<th>Specific heat $C_p$ [J.kg$^{-1}$.K$^{-1}$] (DSC)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wc-Co 9%</td>
<td>$1 \pm 0.01$</td>
<td>$16 \pm 0.01$</td>
<td>$14264 \pm 200$</td>
<td>$41 \pm 4$</td>
<td>$206 \pm 10$</td>
</tr>
<tr>
<td>CuO</td>
<td>-</td>
<td>-</td>
<td>$6300 \pm 120$</td>
<td>-</td>
<td>$460 \pm 10$</td>
</tr>
</tbody>
</table>

*Table 2:* Known thermo-physical properties of the deposit-substrate system.
Table 3: Roughness parameters for substrate and realised CuO films.

<table>
<thead>
<tr>
<th>( R_w ) (nm)</th>
<th>302</th>
<th>286</th>
<th>304</th>
<th>300</th>
<th>281</th>
</tr>
</thead>
<tbody>
<tr>
<td>( R_t ) (nm)</td>
<td>3.6</td>
<td>3.1</td>
<td>3.3</td>
<td>2.7</td>
<td>3</td>
</tr>
</tbody>
</table>
Table 4: Chemical composition (% mass) of sample for 1 µm depth from the deposit surface.

<table>
<thead>
<tr>
<th>Mass%</th>
<th>$e_d=500$ nm</th>
<th>$e_d=750$ nm</th>
<th>$e_d=1000$ nm</th>
<th>$e_d=1580$ nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>6.95 ± 0.56</td>
<td>0.89 ± 0.58</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Co</td>
<td>2.13 ± 0.68</td>
<td>1.10 ± 0.65</td>
<td>0.95 ± 0.57</td>
<td>1.01 ± 0.57</td>
</tr>
<tr>
<td>Cu</td>
<td>78.34 ± 1.50</td>
<td>84.97 ± 1.50</td>
<td>84.98 ± 1.32</td>
<td>85.37 ± 1.32</td>
</tr>
<tr>
<td>O</td>
<td>12.58 ± 0.10</td>
<td>13.05 ± 0.09</td>
<td>14.07 ± 0.08</td>
<td>13.13 ± 0.08</td>
</tr>
<tr>
<td>$e_d$ (nm)</td>
<td>500</td>
<td>750</td>
<td>1000</td>
<td>1580</td>
</tr>
<tr>
<td>-----------</td>
<td>----------</td>
<td>----------</td>
<td>----------</td>
<td>----------</td>
</tr>
<tr>
<td>$\lambda_d$ [W.m$^{-1}$.K$^{-1}$]</td>
<td>0.921 ± 0.014</td>
<td>1.180 ± 0.021</td>
<td>1.449 ± 0.031</td>
<td>1.879 ± 0.033</td>
</tr>
</tbody>
</table>

**Table 5**: Estimated CuO film thermal conductivity according to its thickness.