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To cite this article: A Hamdi et al 2010 J. Phys.: Conf. Ser. 214 012121

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Paraffin oil thermal diffusivity determination using a photothermal deflection setup with a 2.3µm pump: a first step towards methane detection

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Abstract. The photothermal deflection technique, also known as "mirage effect", is a non-destructive method of evaluating thermal properties of solid, liquid or gaseous species. This technique will be used to detect pollutant absorption. As the deflection is stronger in liquids than gases, we will first consider the deflection in paraffin oil. We consider a medium that is heated by a modulated laser diode beam, and we measure the deflection of the probe beam passing through the heated region as a function of the distance between the axes of the beams. After some theoretical considerations and numerical simulations, we present the application of this method to the experimental determination of the thermal diffusivity of a liquid sample in excellent agreement with previously known values.

1. The pump beam: an antimonide laser diode emitting at 2.3µm

In this work, we use as a pump beam a tunable GaSb based laser diode with an active zone constituted of GaInAsSb/AlGaAsSb quantum wells. This device has been grown by molecular beam epitaxy at the Institut d’Électronique du Sud (IES). It works in the continuous-wave regime at room temperature, with an emission wavelength of 2.32µm. This kind of device has already been successfully used in tunable laser diode absorption spectroscopy [1]. The emission spectrum of the pump laser is shown in figure 1 (inset). Figure 1 shows the absorption peaks of methane. This measurement has been performed using a 12mm-long cell filled with pure methane. The laser beam is directed through this cell and detected with an InGaAs photodiode. Around 2.3µm, there is a large methane absorption band (fig 3), constituted of many absorption lines. Tuning the injected current inside a laser diode increases the active zone temperature. The main effect is a variation of the refractive index thus an increase of the emitted wavelength (tuning effect [2]) of the device. It makes it possible to cross any methane absorption lines.
The laser diode shows a threshold current of 110 mA at 25°C with an emission wavelength around 2.32 µm. Its optical power reaches 1 mW at 100 mA with a current tuning of 0.03 nm.mA\(^{-1}\).

As can be seen figure 2, the 2.3µm range is interesting because CH\(_4\) absorption is strong while water absorption is weak [3] in the atmosphere transmission window.

2. Principle of mirage effect detection

When a medium like a liquid or a gas is studied, it is excited by nonuniform absorption of a modulated pump-laser source. The medium heats locally, introducing a locally modulated temperature gradient. This temperature gradient induces in turn a refractive index gradient. Then, a probe beam (here an He-Ne laser at 0.632 µm) passing through this region will be deflected by an angle related to the thermal gradient at the modulation frequency which is related to thermal properties of the medium. The deflection angle is detected by a quadrant silicon photodiode, linked to a lock-in amplifier locked at the pump-laser modulation frequency. The modulation of the pump beam is made with a chopper. Here the main interest of the technique is that an effect, induced by an infrared device (absorption of the pump laser, at 2.3 µm) can be detected by the deflection of a red laser measured by a cheap silicon detector.
3. Theoretical study

The model geometry is shown in Fig.3. Both regions 0 and 2 are optically transparent. Only region 1 will be considered as an absorbing medium. We assume that all regions extend infinitely in the radial direction. We consider the thermal wave propagation in one dimension.

In each region, the heat equation is given by:

\[ \nabla^2 T_i(M,t) - \frac{1}{D_i} \frac{\partial T_i(M,t)}{\partial t} = -\frac{Q_i(M,t)}{k_i} \]  

(1)

Here we only take into account heat transfer by heat conduction governed by Fourier's law. Heat transfer by thermal convection or thermal radiation is neglected.

\( D_i, k_i, Q_i(M,t) \) are the thermal diffusivity, thermal conductivity, and the power density transformed into heat in the \( i \)-th region. \( T_i \) is the temperature rise above the ambient temperature of the \( i \)-th region. Because regions 0 and 2 are transparent, the only source is \( Q_1 \), that is related to the laser intensity distribution function, given by [4]:

\[ Q_1(M,t) = \frac{4}{\pi^2} \frac{P \alpha}{a_p^2} \exp(-\alpha z) \exp\left(-\frac{2r^2}{a_p^2}\right) \exp(i \omega t) \]  

(2)

Here, \( P \) is the optically exciting beam power, \( \alpha \) is the absorption coefficient, \( a_p \) is the radius of the pump beam. The boundary conditions of temperature and heat flow at the different interfaces are:

\[ T_0|_{z=0} = T_1|_{z=0}, \quad T_1|_{z=l} = T_2|_{z=l} \]  

(3a)

\[ k_0 \frac{\partial T_0}{\partial z}|_{z=0} = k_1 \frac{\partial T_1}{\partial z}|_{z=0}, \quad k_1 \frac{\partial T_1}{\partial z}|_{z=l} = k_2 \frac{\partial T_2}{\partial z}|_{z=l} \]  

(3b)

The resolution of equation (1) has been developed in [4]. By resolving the propagation equation of a light beam through a spatially varying refraction index, W.B. Jackson and all ([4] and [5,6]) gives the expression of the deflection angle \( \phi \) as:

\[ \phi = -\frac{1}{2n_0} \frac{\partial n}{\partial T} \int_0^l dz \int_0^\infty \delta^2 J_1(\delta x_0) [\Gamma(\delta)\exp(-\delta z) + A(\delta)\exp(-\beta_1 z) + B(\delta)\exp(\beta_1 z)] d\delta . \]  

(4)

Where \( x_0 \) is the distance between the axes of the two beams.

Here \( J_1(\delta r) \) is the first-order Bessel function of the first kind, and \( \delta \) is an integration variable.

\( A, B, H, \beta_1 \) and \( \Gamma \) were determined by applying the boundary conditions of the temperature and the heat flow in all interfaces, \( z = 0 \) and \( z = l \), we get :

\[ A(\delta) = -[(1 - g)(b - r) \exp(-\alpha l) + (g + r)(1 + b) \exp(\beta_1 l)] \gamma_n(\delta) \]

\[ B(\delta) = -[(1 + g)(b - r) \exp(-\alpha l) + (g + r)(1 - b) \exp(-\beta_1 l)] \gamma_n(\delta) \]  

(5)
\[ H(\delta) = [(1 + g)(1 + b) \exp(\beta_1 l) - (1 - g)(1 - b) \exp(-\beta_1 l) \]
\[ \beta_1^2 = \delta^2 + i \frac{\omega}{D_1}, \quad \gamma_H(\delta) = \frac{\Gamma(\delta)}{H(\delta)} \quad \text{and} \quad \Gamma(\delta) = \frac{\alpha P}{\pi^2 k_1} \frac{\exp[-(\delta a_p)^2/8]}{\beta_1^2 - \alpha^2}. \]

Where: \( g = \frac{k_0}{k_1} \beta_0 \), \( b = \frac{k_3}{k_1} \beta_2 \), \( r = \frac{\alpha}{\beta} \).

4. Experimental setup and results

4.1. Setup
This experimental setup is dedicated to gas detection. We first have made measurements with paraffin oil to build up the setup and calibrate the optical deflection using a strong deflecting medium. The next experimental step will be the replacement of paraffin oil by gaseous methane. The experimental set-up is shown in fig.4. The cell is immersed in air. The pump beam of the laser diode is controlled by a current generator (Model 525 laser diode driver) and thermally regulated with a temperature controller (TED200C). This pump beam is modulated by a mechanical chopper at 9Hz, and focused with an aspherical lens (A220TM, Thorlabs) with focal length 11 mm.

The He–Ne laser probe beam is reflected by a pellicle beam splitter and focused in the cell by a glass lens of focal length 300mm. The deflection of the probe beam is measured by a four quadrants (QD50T) silicon photodetector linked to a lock-in amplifier (EG&G5210: single phase or SR530: double phase).

![Experimental set-up](image)

Fig.4: a: laser diode, b: aspherical lens, c: chopper, d: beam splitter, e: sample, f: photodiode, g: lock-in amplifier, h: He-Ne laser, i: frequency generator, j: glass lens.

4.2. Experimental results
For a fixed value of frequency, the deflection is measured as a function of distance between the pump and probe beams. To determine the thermal diffusivity of the paraffin oil, we have plotted in fig.5 the experimental and theoretical deflection amplitude for three values of the thermal diffusivity (1x10^{-8} m^2.s^{-1}, 5x10^{-8} m^2.s^{-1}, 10x10^{-8} m^2.s^{-1}) as a function of the distance between the axes of the beams.

The value of the diffusivity that overlaps as much as possible the theoretical and experimental curve is \( D = 5 \times 10^{-8} m^2.s^{-1} \). This value is in good agreement with the value of 4x10^{-8} m^2.s^{-1} measured in [7].
At 2.3µm, the paraffin oil absorption coefficient is about 10 cm⁻¹, either the interaction length is about few millimetres. We have used a 10 cm long quartz cell, leading to a total absorption. In the experimental conditions, the noise in the focused configuration does not exceed 10 % the deflection signal. To be able to detect methane absorption, the setup sensitivity has to be increased, for example by improving the deflection detection by using a smaller quadrants detector, by tuning the laser beam through a stronger methane absorption line, and by using a more powerful laser diode.

5. Conclusion
We have shown in these paper preliminary measurements based on mirage effect. Dedicated to gas detection using a 2.3 µm laser diode as a pump laser, the setup has been first developed and characterised with a cell full of paraffin oil, giving a strong absorption and a strong deflection. Using the optical setup in this configuration, we managed to make the theoretical modelization of the deflection and experimental determination of the medium thermal diffusivity with a very good agreement with previous evaluations [7]. The next step of this study is the improvement of the setup sensitivity to apply the mirage effect to gas detection.

References