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Mirage technique in anisotropic solids

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Abstract: Theoretical and experimental analysis of heat diffusion in an anisotropic medium are presented. The solution of the 3D thermal conduction equation in an orthorhombic medium is calculated by the mean of a Fourier transforms method. Experiments were performed on an orthorhombic polydiacetylene single crystal sample. The temperature field at the sample surface was determined using the photothermal probe beam deflection technique. Then the 3 coefficients of the thermal conductivity tensor have been measured.

1. INTRODUCTION

Many theoretical models for thermal diffusion of heat have been reported [1,2], but only a few experimental investigations concerning anisotropic media are available. We can point out the flash method [3] and the "steady-state potentiometric" technique [4] which have been used to analyse thermal properties of orthotropic polymer samples.

For our study, the experimental device used is the photothermal probe beam deflection technique [5, 6]. This method requires no mechanical contact with the sample and thus experiments on very small samples can be performed.

We present a numerical simulation of the probe beam deflection for pure thermal waves progressing in an orthorhombic sample, using the Fourier transform of the generalized 3D thermal conduction equation.

The sample under study is a polydiacetylene single crystal of poly-4BCMU (BCMU for butoxycarbonylmethylurethane). This orthorhombic crystal of conjugated unbranched polymer chains extending in the crystallographic *c* direction [7], is in a (*b*, *c*) planar configuration.

We have determined the 3D thermal conductivity tensor of this sample from the analysis of the tangential component of the deflection angle, versus the offset between the pump and the probe beams.

2. THEORY

2.1 Basic analysis

In the case of an orthorhombic geometry, the second order conductivity tensor can be written as

$$\underline{\underline{\kappa}} = \begin{pmatrix} \kappa_c & 0 & 0 \\ 0 & \kappa_b & 0 \\ 0 & 0 & \kappa_a \end{pmatrix}$$

where κ_c , κ_b denote the thermal conductivity coefficients respectively parallel and perpendicular to the chain direction, and κ_a perpendicular to the (*b*, *c*) plane. Then, according to the energy conservation law, the stationary 3D thermal conduction equation in such a medium, in the reference frame relative to the crystallographic axis (Fig.1) and for a modulation frequency $f = \omega/2\pi$ is :

$$\bar{\kappa} \nabla^2 \tau_s(u, v, z) + i \rho C_p \omega \tau_s(u, v, z) = -\frac{I_0}{2\pi r_g^2} \exp\left(-\frac{u^2 + v^2}{2r_g^2}\right) \exp(\beta z)$$

where I_0 and r_g are respectively the power and the radius of the pump beam. The density, the specific heat and the optical-absorption-rate constant of the sample are denoted by ρ , C_p and β . The right hand term describes the heat source which keeps the cylindrical symmetry of the gaussian pump beam profile; this term is equal to zero in the thermal conduction equation of the surrounding media (respectively τ_g and τ_b for the front and backing air).

FIG. 1. Schematic description of the geometrical configuration. The experimental device is attached to the (X, Y, Z) axis, whereas the (U, V, Z) frame refers to the crystallographic (c, b, a) axes (where c denotes the chain direction). The angle (c, X) is denoted by θ .

In order to obtain the temperature distribution within the sample, we used a 2D Fourier transform in the (u,v) plane, taking account of the boundary conditions at the interfaces (continuity of temperature and heat flow) between the sample and the air. We can consider the sample as infinite in the (u,v) plane, because of its dimensions larger than the characteristic thermal wavelength associated to our frequency modulation range. The temperature distribution τ_g in the probed atmosphere is deduced from the integral in Fourier space :

$$\tau_g(u, v, z) = \int \int_{-\infty}^{+\infty} dk_u dk_v \exp[-i(k_u u + k_v v)] \exp[-\sigma_g(k_u, k_v)z] T_g(k_u, k_v).$$

where $T_g(\mathbf{k})$ is the temperature in the Fourier space [8] and $\sigma_g^2 = k_u^2 + k_v^2 + 2i l_g^{-2}$. In this formula, $l_g = (2 \kappa_g / \rho_g C_g \omega)^{1/2}$ denotes the thermal diffusivity length in the probed air.

The probe beam deflection ϕ is given by the line integral of the temperature gradient along the whole optical path L :

$$\phi = -\frac{1}{n} \frac{dn}{dT} \int_L \nabla \tau_g \times d\mathbf{l}.$$

Then, with the appropriate expression of τ_g in the (X,Y,Z) reference frame, the tangential component ϕ_t is :

$$\phi_t \propto \int_{-\infty}^{+\infty} \frac{\partial \tau_g}{\partial x} dy, \text{ which reduces to : } \phi_t \propto \int_{-\infty}^{+\infty} dk k \sin(kx) T_g(k) \exp[-\sigma_g(k)z] \quad (1)$$

where $\sigma_g^2(k) = k^2 + 2i l_g^{-2}$. Equation (1) is numerically calculated to compare our theoretical model with experimental data.

2.2 Asymptotic derivation

Physical considerations lead to simplifications in the theory presented above :

in our case, the high value of the optical absorption constant ($\beta = 2 \times 10^5 \text{ cm}^{-1}$ compared to a sample thickness $d = 70 \text{ }\mu\text{m}$), and the existence of a mismatch between the densities of the sample (ρ_s) and the probed medium (ρ_g), ($\rho_s \ll \rho_g$), lead to the following assumptions : $\exp(-\beta d) \approx 0$, and $\sigma_g \kappa_g / \sigma_s \kappa_a \approx \sigma_b \kappa_b / \sigma_s \kappa_a \approx 0$;

we introduce an effective thermal conductivity coefficient in the (b,c) plane, relative to the scanning direction x :

$$\kappa_\theta = \kappa_c \cos^2 \theta + \kappa_b \sin^2 \theta \quad (2)$$

and a rescaled sample thickness $\tilde{d} = d \sqrt{\kappa_\theta / \kappa_a}$, equation (1) becomes :

$$\phi_t = -\frac{1}{n} \frac{dn}{dT} \frac{I_0}{\sqrt{\kappa_a \kappa_\theta}} \int_0^\infty dk k \sin(kx) \exp\left[\frac{-r_g^2 k^2}{8}\right] \frac{1}{\sigma_\theta} \frac{1 + \exp(-2\sigma_\theta \tilde{d})}{1 - \exp(-2\sigma_\theta \tilde{d})} \exp(-\sigma_g z) \quad (3)$$

where $\sigma_\theta^2 = k^2 + 2i l_\theta^{-2}$, and $l_\theta = \sqrt{2\kappa_\theta / \rho_s C_s \omega}$ denotes the thermal diffusion length in the direction θ .

Under such conditions, equation (2) has the same form as in an isotropic medium [9].

Furthermore, in the asymptotic limits of a thermally thin ($d \ll l_s$), and of thermally thick ($d \gg l_s$) samples, the integrand is proportional to σ_θ^{-2} and σ_θ^{-1} , and independant of κ_a . So, for low frequency experiments, equation (3) reduces to :

$$\phi_t(x, z) \propto \int_0^\infty dk k \sin(kx) \exp\left(\frac{-r_g^2 k^2}{8}\right) \frac{1}{\sigma_\theta^2} \exp(-\sigma_g z)$$

Under these conditions, it can be shown [8] that the value of κ_θ can be derived from the dependence of the amplitude A_t of ϕ_t in terms of the transverse offset x (for large values of x).

3. EXPERIMENTS

Experiments were performed on a conventional mirage detection setup, which is described in detail in ref. [8]. Its summarized main features are the following: a 8 mW-power argon laser beam, tuned on 488 nm wavelength, provides the polarized gaussian profile pump beam. The scanning of the distance X_0 between the pump and the probe beams is monitored by a 0.1 μm precision step motor. The low-frequency modulation (0.1-20 Hz) is accomplished by an electromechanical shutter (nm Laser Products LS 200).

Monocrystals of poly-4BCMU [10] are obtained by irradiating monomer single crystals with γ rays. These monocrystals are then cleaved along the (b,c) plane and typically 50 to 100 μm thick, 10 mm long and 6 mm wide samples were available. These samples were held by their edges on a Pyrex substrate, with an air layer between the sample and Pyrex holder. The chain direction was found from the strong optical dichroism of the 4BCMU. The maximum absorption of light occurs when the polarization of incident light is parallel to c.

4. RESULTS

Experiments have been performed on samples of different thickness. As the results were the same within experimental error, we present data for only one sample of 70 μm thickness. In Fig. 2 we have plotted the experimental data for $\theta = 90^\circ$ (which means that X_0 is progressing in the perpendicular chain's direction). The curves exhibit a maximum which is closely related to the pump beam radius r_g but does not influence the plot for large values of X_0 . For the low frequency experiments (0.5 and 1 Hz), the slopes of the lines give $\kappa_b = 0.42 \text{ W m}^{-1} \text{ K}^{-1}$. The different behavior of the high-frequency experiments (4 and 10 Hz) allows us to pinpoint the crossover between the thermally thin and thick regimes. A numerical analysis of these higher-frequency measurements, according to Eq.(1), leads to an estimation of $\kappa_a = 0.025 \text{ W m}^{-1} \text{ K}^{-1}$. Computed lines show quite good agreement with experiments.

In the same way, a value of $\kappa_c = 2.2 \text{ W m}^{-1} \text{ K}^{-1}$ can be deduced from low frequency experiments with $\theta = 0$. For intermediate directions, different values of κ_θ derived from the slope of low-frequency

experiments have been found in good agreement with the theoretical dependence of κ_θ with θ , according to Eq.(2) with κ_c and κ_b previously determined.

FIG. 1. Logarithm of the amplitude (A_t) of ϕ_t (for $\theta = 90^\circ$), is plotted for different modulation frequencies (symbols represent experimental data). According to Eq.(1), solid lines were computed for each frequency with the values $r_g = 360 \mu\text{m}$, $d = 70 \mu\text{m}$, $z = 150 \mu\text{m}$, $\rho_s = 1170 \text{ kg m}^{-3}$, $C_s = 1480 \text{ J kg}^{-1} \text{ K}^{-1}$, $\kappa_b = 0.42 \text{ W m}^{-1} \text{ K}^{-1}$ and $\kappa_a = 0.025 \text{ W m}^{-1} \text{ K}^{-1}$.

5. CONCLUSION

We have shown that photothermal-probe-deflection is a suitable technique to study anisotropic thermal conductivity behavior of materials, even when only small samples are available, due to the contactless character of this method. The 3D thermal diffusion equation of heat in an orthorhombic medium was solved using a 2D Fourier transform. So the thermal field in the probed front air can be numerically computed.

The thermal conductivity tensor has been measured by the analysis of the transverse component of the probe beam deflection. We have showed that for optically thick and thermally thin samples, the complete expression reduces to the same one as in an isotropic medium, provided that an effective thermal conductivity relative to the chain direction and a rescaled sample thickness are defined. Moreover, physical considerations lead to simplifications in the theory so that direct measurements of the thermal conductivity were performed.

Various effects may be invoked to explain the large anisotropic behavior of 4BCMU (presence of hydrogens bonds along the chains axis, lamellar structure), but further experimental data (sound velocities, temperature dependence measurements) would be necessary for a complete heat diffusion analysis.

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