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MICROSCALE THERMAL CHARACTERIZATION OF REINFORCED COMPOSITES BY PHOTOTHERMAL MICROSCOPY DATA INVERSION

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Abstract - Within heterogeneous materials like fibber reinforced composite, heat transfer is a complex phenomenon that depends on thermal properties of both fibber and matrix materials and often thermal contact resistance between them. In addition, the fibber axial and radial thermal behaviour is different and both characteristics have to be investigated. In the present work, a photothermal microanalysis method has been used to measure the two values of fibber thermal diffusivity (axial and radial) when inserted inside a matrix. The experimental bench is based on a photothermal method whose periodic excitation is localized on a micron-scale spot. The distance on which the heat has propagated during a period is called thermal diffusion length. When modulation frequencies are higher than 10 kHz, the thermally excited volume does not exceed some μm^3. In this volume, the temperature field is characterized by amplitude and phase - compared to the heating reference - of its periodic component. Local property estimations are deduced from the temperature evolution in a micron-scale zone as well as measured at a given distance. In case of fibber characterization, the composite sample is cut along a plane perpendicular to the fibber axis for measuring the radial diffusivity or containing the fibber axis for measuring the axial diffusivity. With small diameter fibbers, the thermally excited volume is larger than the fibber itself and heat exchange with the matrix have to be taken into account. In this case, the direct problem cannot be solved by an analytical approach; a finite element method (FEM) has been used. Since time dependent solutions are numerically difficult to estimate (for high frequency excitation), a complex temperature is considered and lead to the determination of both phase and modulus in stationary state. Numerical and experimental results are compared and an inversion algorithm based on the FEM is discussed.

1. INTRODUCTION

The developement of new methods capable of characterizing the microscale thermal behavior of heterogeneous complex materials is a crucial step for optimizing elaboration process. In such a way, a thermal diffusivity measuring device has been developed. The experimental bench is based upon a periodic method dedicated to microscopic study. The principle consists in heating a sample by a modulated laser beam on one face and to measure a temperature dependent coefficient (reflectance) near the heating source. The temperature evolution (induced by the periodic heating input) is the sum of a continuous component and a periodic one characterised by its amplitude and phase lag versus thermal excitation. Reflectance being temperature dependent, the phase lag between reflectance variations and heating input depends on the thermal properties of the heated volume. Thus, it leads to the identification of the thermal diffusivity of the material in a microscopic area ($\leq 20\mu m$). This experimental bench is developed in order to study at a microscopic scale :

- thermal characteristics of materials (thermal property distribution, anisotropy, …),
- thermal interfaces between multicomponent materials,
- thermal discontinuities (cracks, …) .

An inverse problem has to be solved in order to identify the thermal diffusivity of the studied material. Minimization of difference between observed phase lag and simulated phase lag is performed. Simulated values are deduced from a mathematical model describing the heat transfer induced by periodic excitation. Semi analytical solutions are proposed in [7] for homogeneous samples and in [11] for several types of discontinuities. In both previous communications, the partial differential equation system is solved using a space Fourier transform; calculation of the inverse Fourier transform is carried out numerically and gives the temperature amplitude and phase lag values compared to the incident heat flux. However, the semi-analytical solution validity sharply depends on strong hypothesis which can be quite difficult to verify for heterogeneous materials. In fact, the distance on which the heat has propagated during a period is called thermal diffusion length $\delta = \sqrt{a/\pi f}$, where a is the thermal diffusivity and f is the modulation frequency. In the studied configuration, the thermally excited volume does not exceed some μm^3. When spatial heterogeneities dimension is about the thermal diffusion length (see Table 1), the semi-analytical solution is not valid. For example, identification of thermal diffusivity in micrometric fibers whose diameter is less than $10\mu m$, can not be performed using inverse Fourier transform. In such a framework, a numerical solution based on the FEM is proposed [2]. The measuring bench and experimental results are shown in Table 1.

Frequency $[Hz]$	diffusivity $\left[m^2.s^{-1}\right]$	diffusion length $[\mu m]$
$f=10^4$	$a=10^{-5}$	$\delta \approx 20$
$f=10^6$	$a=10^{-6}$	$\delta \approx 0.5$

Table 1. Diffusion length estimation.

2. MODELING IN THE FREQUENCY DOMAIN

Thermal waves produced by a periodic heat generation in homogeneous and inhomogeneous solids are examined from the theoretical point of view in [8]. Application to thermal diffusivity measurement using harmonic and one-dimensional propagation of thermal waves is proposed in [10]. Let us consider the following notations : $\Omega_i \subset \mathbb{R}^3$ is the space domain corresponding to the component i, $X=(x,y,z)\in \bigcup\Omega_i$ is the space variable, $t\in[0,T]$ is the time variable. In [11], the periodic heat flux focused at the surface Γ on point I is expressed in the form :

$$\phi(r_X,t)=\phi_0 e^{-\frac{r_X^2}{r_0^2}} e^{j\omega t} \tag{1}$$

where ϕ_0 is the heat flux amplitude $\left[W.m^{-2}\right]$, r_X is the distance XI in $[m]$, r_0 is characteristic of the heat flux spatial distribution $[m]$, ω is the pulsation $\left[rad.s^{-1}\right]$. The evolution of temperature $\theta(X,t)$ in $\bigcup\Omega_i$ is described by, the following equations :

$$\forall(X,t)\in\bigcup\Omega_i\times(0,T] \qquad \Delta\theta(X,t)-\frac{1}{\alpha_i}\frac{\partial\theta(X,t)}{\partial t}=0 \tag{2}$$

where α_i is the unknown diffusivity,

$$\forall(X,t)\in\Gamma\times(0,T] \qquad -\lambda_i\frac{\partial\theta(X,t)}{\partial\vec{n}}=\phi(r_X,t)-h\theta(X,t) \tag{3}$$

where λ_i is the thermal conductivity, $\vec{n}$ is the normal vector exterior to Γ, h is the convective exchange coefficient,

$$\forall X\in\bigcup\bar{\Omega}_i \qquad \theta(X,0)=0. \tag{4}$$

Since the heat flux is periodic on Γ, temperature variations in $\bigcup\Omega_i$ will be periodic as well. When the steady-state is established, a continuous component and a periodic one are considered :

$$\theta(X,t)=\theta_c(X)+\theta_\omega(X)e^{j\omega t}. \tag{5}$$

In the following, the study is devoted to the periodic component, i.e. computation of its amplitude and phase lag with respect to the incident flux. From (2)-(4), we obtain :

$$\forall X\in\bigcup\Omega_i \qquad \Delta\theta_\omega(X)-\frac{j\omega}{\alpha_i}\theta_\omega(X)=0 \tag{6}$$

$$\forall X\in\Gamma \qquad -\lambda_i\frac{\partial\theta_\omega(X)}{\partial\vec{n}}=\phi(r_X)-h\theta_\omega(X) \tag{7a}$$

$$\forall X\in\bigcup\bar{\Omega}_i \qquad \theta_c(X)=-\theta_\omega(X) \tag{7b}$$

where $\phi(r_X)=\phi_0 e^{-\frac{r_X^2}{r_0^2}}$.

In specific configurations such as homogeneous solid, semi infinite geometries, temperature independent parameters, particular multi-component configurations (for which thermal interfaces are well identified), calculation of the inverse Fourier transform leads to a semi-analytical solution, see theoretical aspects and applications in [5], [6], [7], [9] and [11]. From the experimental point of view, for heterogeneous materials which do not verify previous assumptions, thermal diffusivity identification according to semi-analytical solution can lead to erroneous estimation. In order to provide a general alternative for the resolution of eqns (6) and (7), the FEM is implemented. The complex temperature $\theta_\omega(X)$ is separated in real part and imaginary part : $\theta_\omega(X)=\theta_{\mathrm{Re}}(\omega,X)+j\theta_{\mathrm{Im}}(\omega,X)$. Then, the following coupled systems have to be solved :

$$\begin{cases} \Delta\theta_{\mathrm{Re}}(\omega, X) = -\dfrac{\omega}{\alpha_i}\theta_{\mathrm{Im}}(\omega, X) & \forall X \in \bigcup\Omega_i \\ -\lambda_i \dfrac{\partial\theta_{\mathrm{Re}}(\omega, X)}{\partial\vec{n}} = \phi(r_X) - h\theta_{\mathrm{Re}}(\omega, X) & \forall X \in \Gamma \end{cases} \qquad \begin{cases} \Delta\theta_{\mathrm{Im}}(\omega, X) = \dfrac{\omega}{\alpha_i}\theta_{\mathrm{Re}}(\omega, X) & \forall X \in \bigcup\Omega_i \\ -\lambda_i \dfrac{\partial\theta_{\mathrm{Im}}(\omega, X)}{\partial\vec{n}} = -h\theta_{\mathrm{Im}}(\omega, X) & \forall X \in \Gamma \end{cases} \tag{8}$$

An iterative procedure is carried out in order to solve these systems.

3. SIMULATION AND NUMERICAL RESULT

In the following, micrometer fibers are investigated; transversal and radial thermal diffusivity have to be identified. Several geometrical configurations are considered (see Figures 1 and 2).

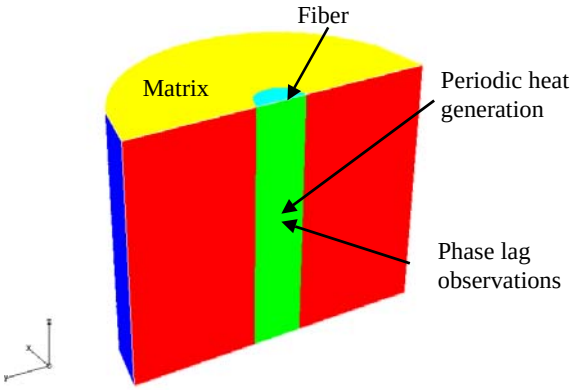

Figure 1. Geometrical configuration for transversal identification.

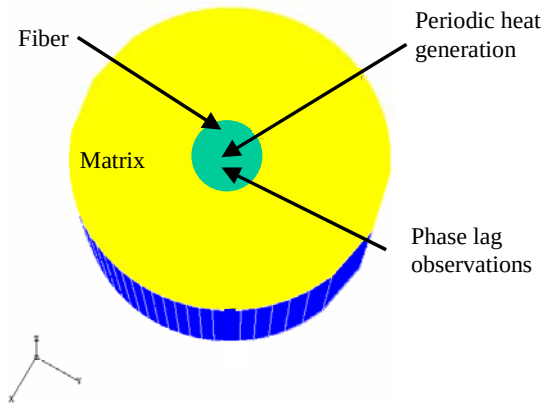

Figure 2. Geometrical configuration for radial identification.

The corresponding meshes are shown in Figure 3. Fibers diameter is about $8\mu m$. Meshes are adapted in the neighbourhood of point I where the periodic heating flux is focused at the surface Γ and near the phase lag observation point.

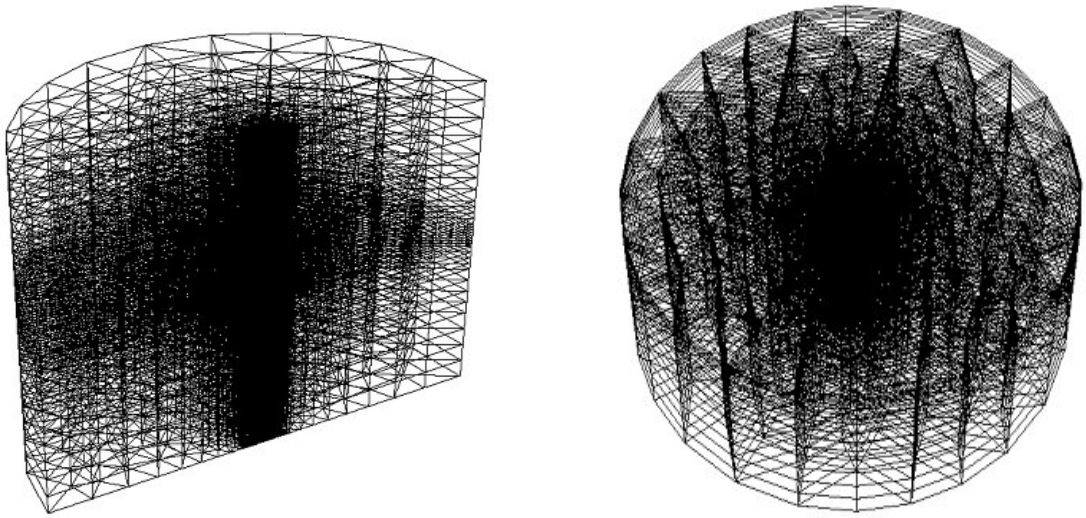

Figure 3. Finite element meshes.

The systems (8) are solved according to an iterative procedure. According to the studied configuration, the phase lag is determined and an example of a relevant result is shown in Figure 4 for a given fiber diffusivity.

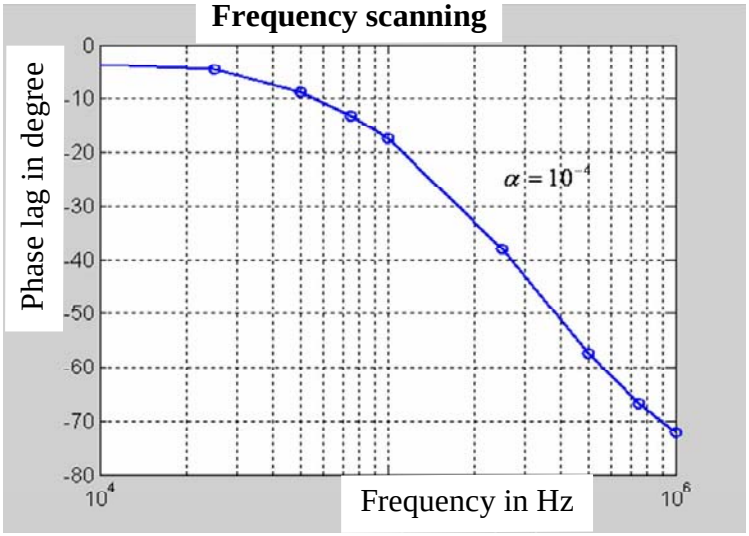

Figure 4. Example of simulation results obtained according to the FEM.

In the following section, the microscopic measurement bench is exposed.

4. MEASURING BENCH

The experimental device used for obtaining measurements able to characterize the microscale thermal behavior of heterogeneous materials is a versatile photothermal microscope. Although the principle of such device is well-known since Rosencwaig *et al*. [12] it will be reminded in order to point out its main advantages and drawbacks.

4.1 Description

The measurement technique is based on the sample thermal response when it is submitted to a microscale periodic thermal excitation. A modulated laser beam (pump), focused by a microscope objective onto the sample surface, produces a local thermal excitation (≈ 1 µm diameter spot). At a given distance (≈ 4-5 µm), a continuous laser beam (probe) is used to detect the thermal wave diffusion by observing the variations of the surface reflectance that depends on temperature [4], [6] and [9]. In our experiment (see Figure 5), the thermal excitation is delivered by an ion-argon laser (COHERENT, Innova 305), the 514 nm waveband of which being selected.

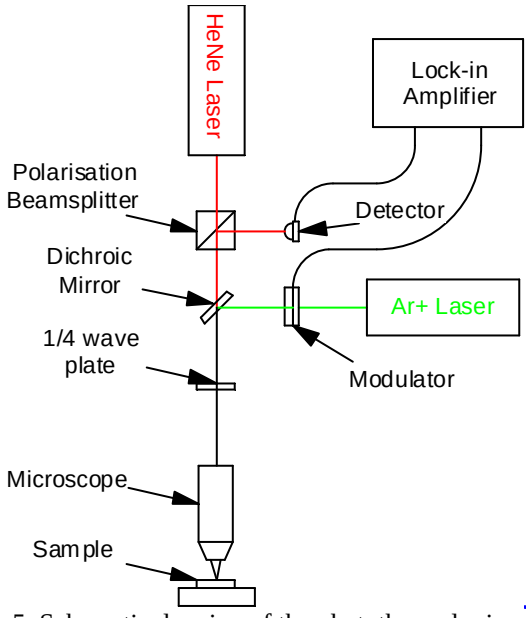

Figure 5. Schematic drawing of the photothermal microscope.

An acousto-optic modulator (ISOMET 1211) driven by a computer-programmable function generator and a RF amplifier modulate the beam at the desired frequency. After shape setting, the beam is reflected by a dichroic plate and focused by a microscope objective (x50, MITUTUYO) on a gaussian microscale spot at the sample

surface. The 632 nm measurement beam, originated from a He-Ne laser (ORIEL 79200) crosses a polarization beamsplitter and the dichroic plate, then is directed to the same objective which focuses it close to the heating spot. The distance between spots (called offset) is accurately adjusted by means of wedge prisms rotation. The reflected part is sent back to the polarization beamsplitter which reflects it towards a fast response photodiode. The photodiode AC component signal is amplified and analyzed by a wide bandwidth lock-in device (EGG 5302), the reference signal of which comes from the acousto-optic controller. The lock-in amplifier output (amplitude and phase lag) is finally recorded by the control computer.

The phase lag between reflectance variations and heating laser modulation corresponds to the thermal diffusion process between excitation (pump) and observation (probe) spots. The unknown thermophysical properties are thus microscale characteristics of the investigated zone. These properties are then identified by analyzing the evolution of the phase lag versus an adjustable parameter (independent variable) such as the excitation frequency, distance between spots or distance from a thermal discontinuity.

4.2 Calibration procedure

The experimental system (optical and electronic devices) introduces an additional phase shift that should be subtracted from the measured value to keep only the thermal contribution. For this, a calibration procedure consists in picking up with an optical fiber a small part of the pump beam and sending it directly to the detector. The resulting phase, measured on the whole frequency range, is stored in a table that will be used to correct the set of experimental values obtained on the samples.

Because of the short distance between pump and probe spots, their shape and size are to be taken into account. Although commercially available beam analyzers do allow measurements of microscale beams, they are not adapted for analyzing beams focused by such high numerical aperture objectives. The spots characteristics are investigated by a two-step procedure [5]. A scanning slit beam profiler (DATA RAY Beamscope P5) capable of measuring focused beam profiles of some tens of microns is used to analyze the beam shape in several locations upstream and downstream the waist. This step allows to verify that the beams are gaussians and to determine the beam quality coefficient (M^2). It becomes then possible to extrapolate the value of the waist diameter by applying the gaussian beam propagation law.

Analysis performed on the system equipped with the specified objective (x50, MITUTUYO) gives the following results :

- pump spot diameter: 1.00 ± 0.04 µm,
- probe spot diameter: 1.24 ± 0.04 µm.

4.3 Sample holder and positionning

The sample is held by a two-stage micropositioning system (0.1 µm resolution) driven by the computer. This allows 1-D scanning of the sample surface that will be used for thermal parameter estimation, or 2-D scanning for imaging the map of surface thermal transfers. Heterogeneous samples often comprise materials of different hardness and so the polishing results in surface altitude variations of some microns. Because of the low depth-of-field, these variations have to be corrected. The sample holder involves a third movement in the z-direction in order to maximize the probe laser reflected beam.

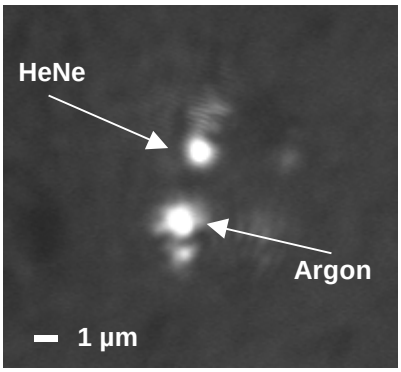

Figure 6. Example of surface image.

A CCD camera sights the surface by means of a beamsplitter. Its aim is an accurate positioning of both spots on the sample surface as well as a measurement of the distance between spots. (Note: disturbing reflections observed on Figure 6 come from color filters and not from the surface). The various illumination levels (pump and probe spots, surface lighting) are very different and so a raw image would be saturated and unusable. In order to balance them, the surface is lightened by a pulsed 932 nm diode and the composite beam reflected by the surface passes trough color filters attenuating the 514 nm and 632 nm wavebands.

5. EXPERIMENTAL RESULTS

The measuring device has been used for several fiber configurations (see Figures 7 and 8).

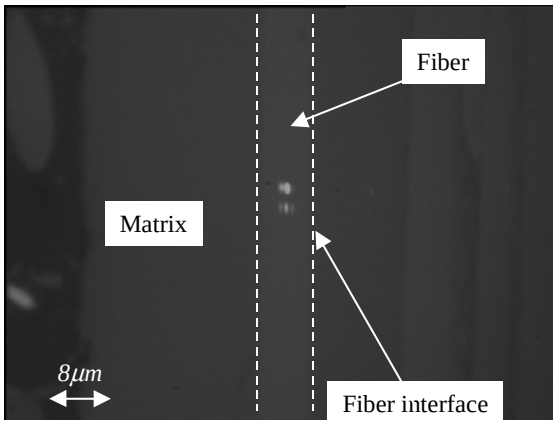

Figure 7. Measurements for transversal identification

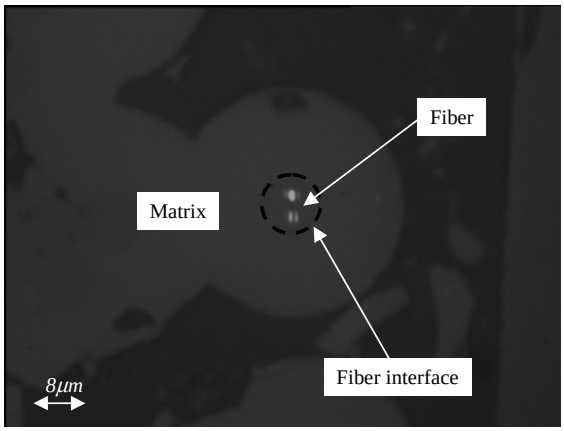

Figure 8. Measurements for radial identification

Examples of phase lag measurements are shown in Figure 9. Comparisons between simulation in Figure 4 and observations in Figure 9 seems to ensure that :

- thermal model described by the coupled systems (8) in order to determine the real part and imaginary part of θ_ω is well adapted to phase lag estimation,
- experimental bench leads to phase lag measurements in microscale heterogeneous materials.

An identification procedure has to be developed in order to estimate thermal diffusivity by minimizing the difference between simulated phase lag (Figure 4) and measured phase lag (Figure 9). For the resolution of such an inverse problem, one can refer to [1], [3] and [13].

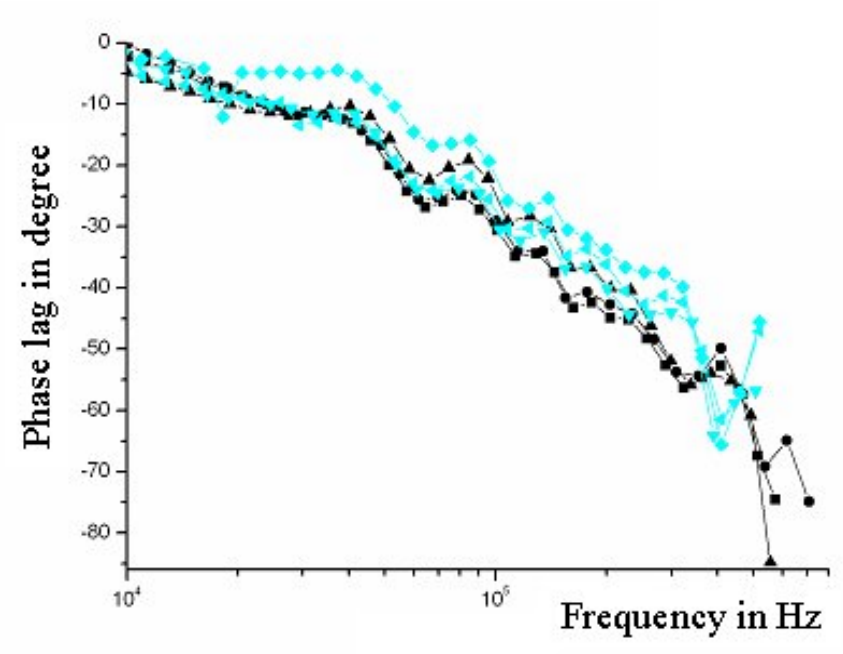

Figure 9. Example of phase lag measurements.

6. CONCLUDING REMARKS

In this communication the interest of a finite element approach in parametric identification is presented. An experimental bench (dedicated to microscopic scale study) is developed for the identification of thermal diffusivity in a heterogeneous material. To estimate this property a model is presented and resolution by the FEM finite element method is proposed in order to improve the identification when semi-analytical solutions can not be considered.

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