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Nonlinear fundamental photothermal response: experimental results for tungsten

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Abstract

A quantitative analysis based on a new three-dimensional nonlinear theoretical model of the experimental fundamental nonlinear photothermal response from multilayer structures containing a tungsten layer is presented. Two sets of wafers, one with a very rough tungsten overlayer surface and another with a smooth polished tungsten surface have been studied. It is shown that the rough tungsten surface gives rise to a very strong nonlinear behavior of the thermal-wave field. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Over the past years, growing interest in nonlinear photothermal phenomena has been motivated by several experimental studies which demonstrated that thermal-wave second harmonic detection can provide better contrast in photothermal microscopy [1,2]. Rajakarunanayake and Wickramasinghe [1] first described nonlinear photothermal deflection imaging experimentally, where the pump beam is modulated at angular frequency ω and the signal is detected at 2ω. Wetsel and Spicer [2] also demonstrated the nonlinear effect in photothermal deflection imaging. Wang and Li [3] further developed a photothermal inspection technique using the photothermal infrared radiometric scheme to detect the second-harmonic response. Finally, Marinelli et al. [4] studied nonlinear heat transport processes in the liquid crystal.

The common feature of all the previous studies is that only the second harmonic detection has been used, while no experimental works have addressed the problem of the fundamental nonlinear photothermal response. Yet, the development of such a theoretical and experimental framework is of significant practical interest for the nondestructive evaluation (NDE) studies of materials exhibiting a strong nonlinear behavior.

In this work we report the quantitative analysis of the experimental three-dimensional fundamental photothermal responses from tungsten wafers using a general three-dimensional nonlinear theoretical model [5].

2. Experimental

2.1. Samples

Two sets of multilayer structures containing a tungsten overlayer of various thicknesses have been studied. The first set (called W1) consisted of seven wafers with the tungsten layer thickness ranging from 3000 to 5800 Å. The wafers from this W1 set have a very rough tungsten surface as confirmed by optical analysis using a microscope. The second set (called W2) consisted of the wafers with tungsten overlayer thicknesses from 3000 to 4500 Å. In contrast to the W1 set, the wafers from this set have a very smooth tungsten surface.

2.2. Experimental set up

Experimental results for the present work were obtained by using a new photothermal system [6] based on the

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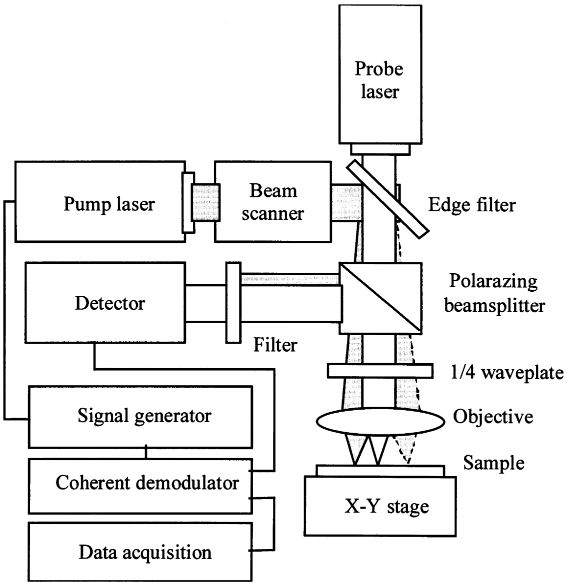

Fig. 1. Block diagram of the experimental set up featuring the pump–probe beam offset scanning block.

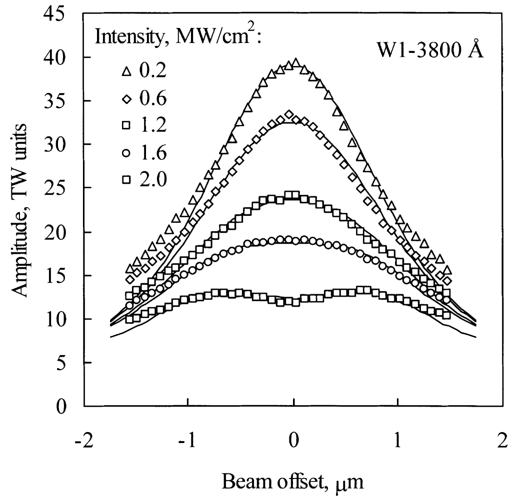

Fig. 3. Experimental fundamental thermal-wave amplitude profiles obtained for 3800 Å tungsten layer from the W1 set at different pump beam intensities (points), and the results of fitting to the model (lines). Other parameters are the same as in Fig. 2.

photomodulated thermoreflectance technique [7] and employing the lateral probing of the thermal-wave field by scanning the pump–probe beam offset distance. Schematic presentation of the experimental set up is shown in Fig. 1. An analysis of the new photothermal system's capabilities in measuring the thickness and the thermal parameters of single metal structures and of multilayer stacks containing Al, Ti, TiN, and SiO_2 layers has been presented elsewhere [8].

In the present work the thermal-wave lateral scans have been measured at several different intensities of the pump beam (diameter 1 μm) at the modulation frequency of 1 MHz. The pump–probe beam wavelengths have been set to 0.79 and 0.67 μm, respectively. The pump–probe beam offset (defined as a distance between the centers of the pump and probe beam spots on the sample surface) has been varied between −1.5 and 1.5 μm.

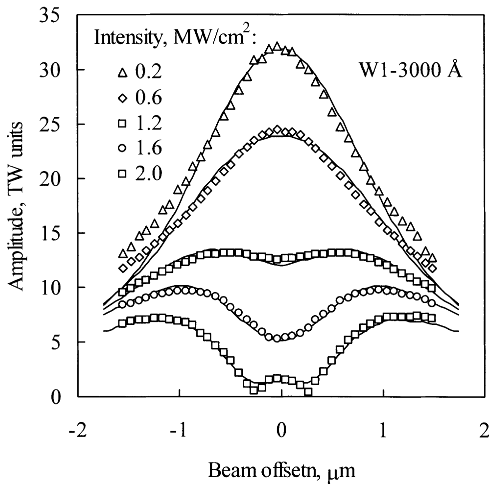

Fig. 2. Experimental fundamental thermal-wave amplitude profiles (in TW units proportional to change in reflectivity $\Delta R/R$) obtained for 3000 Å tungsten layer from the W1 (rough surface) set at different pump beam intensities (points), and the results of fitting to the model (lines). Pump and probe beam diameters: 1 μm, modulation frequency: 1 MHz.

3. Results and discussion

3.1. Rough-surface tungsten wafers

The wafers from the first set (W1) with a rough tungsten surface have been found to exhibit a very strong nonlinear behavior in the pump beam intensity range of 0.2–2.0 MW/cm^2.

Fig. 2 shows the experimental lateral scans of the thermal-wave field obtained from the wafer with the thinnest tungsten layer of 3000 Å. As can be seen from this figure, the nonlinearities introduced by the thin tungsten overlayer result in dramatic changes in the shape of amplitude lateral scans exhibiting a characteristic wave-like behavior as predicted by the theoretical model [5].

Experimental lateral scans have been fitted to the theoretical model in which the nonlinear specific heat $C(T)$ and thermal conductivity $k(T)$ are defined as follows:

$$C(T) \equiv C_0[1 + \delta_1 T(\vec{r}, t)] \quad (1)$$

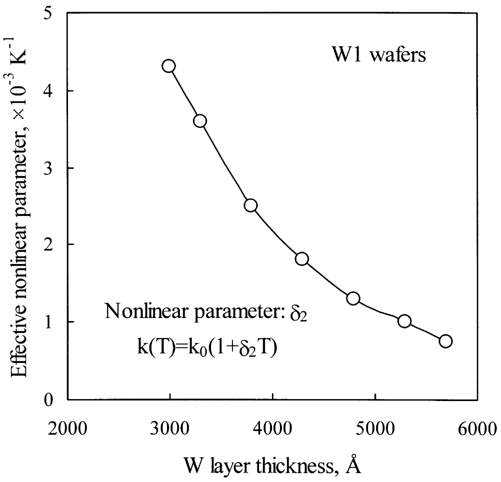

Fig. 4. Effective nonlinear parameter δ_2 as a function of tungsten layer thickness obtained by fitting of the experimental beam offset scans measured on wafers from the W1 (rough surface) set.

and

$$k(T) \equiv k_0[1 + \delta_2 T(\vec{r}, t)] \quad (2)$$

where C_0 and k_0 are the linear specific heat and thermal conductivity, respectively, and δ_1 and δ_2 are the corresponding effective nonlinear parameters [5].

The results of the best fit to the theoretical model are shown in Fig. 2. All amplitude lateral scans recorded at different pump beam intensities have been fitted simultaneously using the general Eq. (51) of Ref. [5] and the same set of constant fitting parameters: $\alpha_0 = 0.69\ \mathrm{cm^2/s}$, $\rho C_0 = 2.52\ \mathrm{J/cm^3\ K}$, $k_0 = 1.73\ \mathrm{W/cm\ K}$ and $L = 1000$ Å,

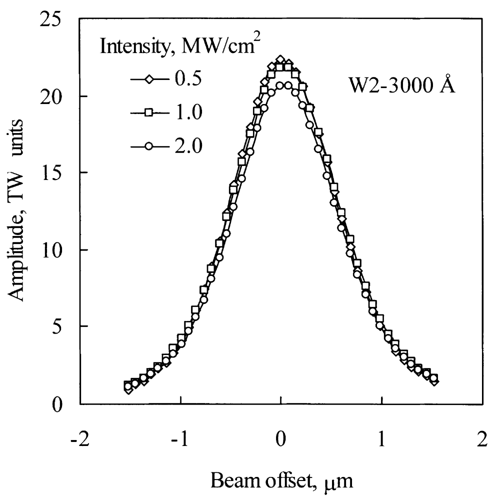

Fig. 5. Experimental thermal-wave amplitude profiles obtained for 3000 Å tungsten layer from the W2 set at different pump beam intensities (points), and the results of fitting to the model (lines). Other parameters are the same as in Fig. 2.

where α_0, ρ and L are the thermal diffusivity, density and the nonlinear layer thickness, respectively. The only variable parameters were δ_1 and δ_2.

As can be seen, the experimental and theoretical results are in very good agreement. It has been found that the nonlinear behavior observed for this sample is mostly due to the nonlinear thermal conductivity of the upper tungsten layer. Fitting revealed $\delta_2 = 4.5 \times 10^{-3}\ \mathrm{K^{-1}}$ and $\delta_1 = 2 \times 10^{-5}\ \mathrm{K^{-1}}$. The decrease in the magnitude of the fundamental photothermal response with increasing laser fluence is due to the increasing channeling of photothermal energy into higher harmonics.

Fig. 3 shows another example of a very good agreement between the theory and experiment observed for another tungsten sample (3800 Å) from the W1 set with the same degree of roughness. However, the nonlinear effect for the wafer with the thicker tungsten layer is lower than that for the sample shown in Fig. 2. In this case fitting revealed a lower thermal conductivity nonlinear parameter $\delta_2 = 3.6 \times 10^{-3}\ \mathrm{K^{-1}}$.

The same nonlinear behavior of the thermal-wave field with decreasing effective nonlinear parameter δ_2 (and practically unchanged δ_1) was observed for the rest of the wafers from the W1 set (Fig. 4).

3.2. Smooth-surface tungsten wafers

In contrast to the previous case, the smooth-surface wafers from the W2 set with tungsten layer thicknesses in the same range do not show any significant nonlinear behavior.

Fig. 5 shows the experimental amplitude lateral scans obtained from one of the wafers from the W2 set. It can be seen that even in the case of the thinnest tungsten overlayer (3000 Å) that was supposed to exhibit the strongest nonlinear behavior, the perturbation in the shape of the lateral scans measured at different pump beam intensities is very small.

Fitting to the theoretical model performed in the same way as for the W1-wafers revealed a very low nonlinear parameter δ_2 on the order of $\sim 10^{-5}\ \mathrm{K^{-1}}$, i.e. two orders of magnitude less than for the rough-surface sample with the same tungsten layer thickness as the W1 set.

Analysis of the data from the W1 and W2 sets of wafers allowed us to hypothesize that the nonlinear behavior observed for the rough-surface tungsten wafers is due to a very thin homogeneous overlayer formed by the tungsten surface roughness and the air interlayer, characterized by a very strong nonlinear thermal conductivity and activated by strong temperature gradients across its thickness. Further work will be required to elucidate the geometrical and thermophysical properties of this highly nonlinear rough overlayer.

Although more extensive experimentation is doubtlessly required to resolve the details of the distinctly different nonlinear behavior observed with the films with rough and smooth surfaces, a physical picture can be constructed which satisfies all our observations, at least phenomenologically.

The rough opaque surface layer, with roughness being hypothesized as pyramids or asperities at possibly quite elevated temperature (thermal-wave magnitude) due to efficient thermal confinement, receives the laser heating and is very likely to exhibit considerable thermal-wave power localization compared to that of the underlayer (substrate). Therefore, the surface magnitude of the thermal wave is expected to be controlled by the roughness and independent of the tungsten film thickness, which is otherwise totally thermally thin at the frequencies of our experiments.

Owing to its huge (physical and thermal) mass compared to that of the thin tungsten layer, the substrate acts as a heat sink for all tungsten thicknesses. Thus, it is reasonable to assume that it is the thermally thick substrate that controls the boundary temperature. With the W–Si interface temperature fixed at constant laser irradiance for all tungsten film thicknesses, and with the surface temperature magnitude fixed by the rough layer geometry, the gradient dT/dz (z: depth coordinate) is expected to be steeper for a thinner film than for a thicker one.

Since the thermal conductivity is a linear function of the temperature elevation above its (dc) room-temperature value, k_0, it is immediately evident that dk/dT will be larger across the thinner tungsten film: a greater $k(T)$ variation will exist across the body of the tungsten film within an element dz at any depth z, due to the larger dT increment across dz. All variations in the thermal-wave magnitude $T(z)$ are appropriately accounted for by the theoretical model, so Fig. 4 is a statement of the greater variance of the $k(T)$ value for larger temperature excursions integrated over the thickness of the upper layer.

In juxtaposition to this situation, when there is little or no roughness on the tungsten layer to "pin" the thermal-wave magnitude, the smooth (and thermally very thin) tungsten layer is expected to perform thermal oscillations essentially isothermally across its thickness, regardless of the actual value of the thickness. Since the substrate still controls the W–Si interface temperature due to its much higher thermal and physical mass, the temperature gradients across all tungsten films are expected to be very weak (near-isothermal case), resulting in the observed insensitivity of δ_2 to the layer thickness and to laser fluence. In other words $\delta_2 = (1/k_0)dk/dT \sim 0$, which renders the tungsten film and the entire two-layer structure conductively linear, as observed in Fig. 5.

δ_1 does not seem to affect the data to any large extent, although dC/dT of the tungsten layer is fully expected to exhibit non-linearities for the steep temperature profile in the rough tungsten case commensurate to the dk/dT gradient. This experimental fact is related to the way in which the thermal-wave signal from the overlayer is generated. In Ref. [5] it was shown that the thermal-wave amplitude depends only on $1/k_1$ and not on C_1. Therefore, only nonlinearities in the thermal conductivity of the overlayer will affect the thermal-wave (ac) signal. The nonlinearities in $C(T)$ show up mostly in the accumulated dc signal level, essentially moving it rigidly up and down. This signal component, however, is filtered out by the lock-in amplifier and cannot be measured in our experiments.

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