Photothermal measurement of absorption and scattering losses in thin films excited by surface plasmons

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We present a novel noncontact, photothermal technique, based on the focus error signal of a commercial CD pickup head that allows direct determination of absorption in thin films. Combined with extinction methods, this technique yields the scattering contribution to the losses. Surface plasmon polaritons are excited using the Kretschmann configuration in thin Au films of varying thickness. By measuring the extinction and absorption simultaneously, it is shown that dielectric constants and thickness retrieval leads to inconsistencies if the model does not account for scattering. © 2009 Optical Society of America

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Surface plasmon resonance spectroscopy has become a very powerful experimental technique that may be used to characterize optical constants of thin films or as a potential biosensor [1–3]. Experimental reflection curves are usually fitted to a three- or four-layer model (Fresnel equations) that does not contemplate scattering. Since results are obtained from curve fitting, these techniques are very model dependent [1,2]. So far, no theoretical models taking into account scattering losses have been proposed. Therefore, scattering is usually disregarded for practical reasons, to the detriment of the accuracy of the retrieval of parameters. Effectively measuring scattering losses in thin films might shed light on some of the discrepancies shown between experimental results and models.

A photoacoustic method was used by Inagaki and coworkers [4] to measure absorption losses. They were able to infer that other radiative losses should be present to explain the attenuation in the reflection curve. A few years later, Barnes and Sambles [5] effectively measured scattering and reflection curves demonstrating the presence of an angle-dependent scattering. Decay mechanisms of surface plasmons in thin films were later studied using techniques such as attenuated total reflection, forward scattering, and combined photoacoustic attenuated total reflection [6].

In this Letter we show a noncontact, photothermal method capable of characterizing the absorption of any nanostructure by measuring—with a lateral resolution of $\approx 0.5\ \mu$m and an axial resolution of ≈ 10 pm—the thermal expansion of a glass microscope coverslip that is in thermal contact with the structure. This allows us to characterize absorption and then calculate scattering losses. In particular, we show experimental results using Au thin films where surface plasmon polaritons (SPP) are excited using a Kretschmann configuration.

The experimental setup consists of a displacement sensor mounted on a commercial total internal reflection microscope (TIRM, Olympus Model IX71) (see Fig. 1). Thin Au films of various thicknesses are sputtered on microscope coverslips. The samples are excited by a doubled Nd:YAG laser at 532 nm (Coherent Verdi) modulated at a frequency of 6.3 kHz using an acousto-optic modulator (AOM) and with a maximum optical power output of ≈40 mW. Photodiode 1 is used to monitor the pump laser power. A *p*-polarized incident beam is selected in order to successfully excite SPP modes. The beam reaches the sample collimated by focusing with lens L1 on the back focal plane of a 1.45 NA, 60×, oil immersion objective. The angle with which the incident beam strikes the sample is controlled by changing the point on which the beam is focused using a galvoscanner. This angle can exceed the total internal reflection angle. Photodiode 2 measures the reflection at the glass–film interface.

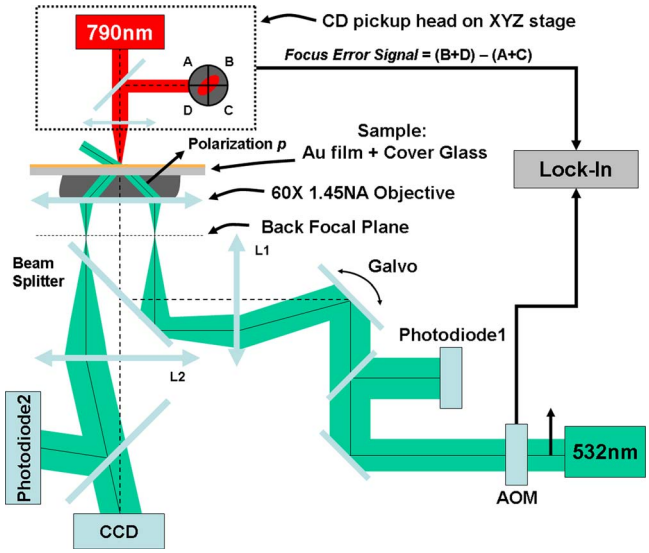

Fig. 1. (Color online) Experimental setup: a TIRM with a CD pickup head mounted on top as a photothermal detector. L1, L2, and the beam splitter are part of the commercial TIRM.

A commercial CD pickup head and its corresponding electronics (LG GCR-8525B CD drive with a Sanyo DL-3150-101 790 nm laser diode) are used to sense the photothermal expansion in this experiment. The pickup reads the information off a CD with a 790 nm IR laser focused on the reflective surface of the CD. The so-called focus error output (FEO) signal is obtained by impinging with the astigmatic IR beam on a four-quadrant detector after reflecting on the surface of the CD. To keep the CD in focus there is a feedback system that monitors changes in FEO and supplies current to a pair of coils that move the objective lens inside the pickup head. The FEO signal is proportional to the distance between the focal plane of the IR laser and the reflective surface of the sample. Thus, by monitoring the FEO signal, we can measure the movement of any reflective surface [7].

To increase the system's sensitivity to motion, we modulate the pump laser beam and perform lock-in detection of the FEO signal. By doing so, we are able to achieve a sensitivity of ≈ 1 pm/μV with a linear range of ≈ 7 μm. Alignment of both laser beams—the pump at 532 nm and the probe at 790 nm—is made possible by using a CCD camera located at the side port of the microscope. It is important to be certain that changes in the FEO signal are due only to thermal expansion. By centering both beams using the CCD camera, we eliminate any undesired contribution from photodeflection of the IR beam [8]. Accordingly, photoreflectance effects can be neglected by locating the sample in the focal plane of the IR laser. With the described experimental scheme, reflection of the pump beam and thermal expansion of the glass coverslip due to light absorption of the sample can be measured simultaneously as a function of the incidence angle.

A schematic analysis of light interacting with a thin metal film is shown in Fig. 2(a). An evanescent wave is generated in the glass–metal interface by exciting the sample with an angle of incidence that exceeds the angle of total reflection. These evanescent waves are responsible for exciting SPP modes that propagate on the metal–air surface when p polarization is used. Of the incident light power, part is reflected back by the glass–metal interface and collected by the microscope objective. Since we are interested in working at angles of incidence beyond total reflection, where no light is transmitted, the rest of the light power is either absorbed in the Au film or scattered by its surface. Absorption depends on the complex part of the dielectric function of the metal, while scattering losses are associated primarily with surface roughness.

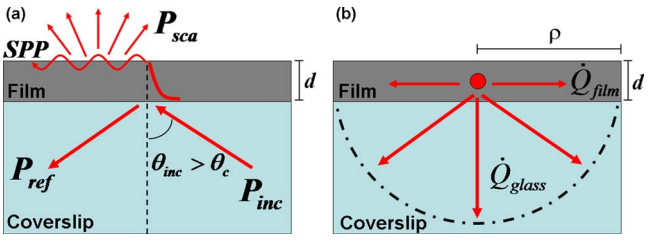

Fig. 2. (Color online) (a) Analysis of light interaction with the sample. A beam strikes the sample with an angle beyond the critical angle, thus generating an evanescent wave that penetrates the film. This evanescent wave excites a surface plasmon. Scattering takes place on the surface of the thin metal film. (b) Heat-flow analysis for a thin film considering a Gaussian heat source.

In our experiment, we measure the reflected and absorbed power and then calculate the losses due to scattering. Thermal expansion of the glass substrate due to the heating produced by the Au thin film, δL_z, can be obtained by solving the heat diffusion equation. For a Gaussian heat source modulated at frequency ω [8,9] we obtain $\delta L_z|_\omega = \Gamma P_{abs}$, where P_{abs} is the power absorbed by the Au film and Γ is the proportionality constant (function of the thermal properties of the glass substrate). To implement this thermal expansion model, we must be able to neglect the lateral heat flow along the metal film, when compared with that inside the glass. If this were not the case, part of the absorbed light power would not contribute to the thermal expansion of the glass and the proportionality hypothesis would not be correct. We test the situation by analyzing the heat flow in both the metal thin film and the glass substrate [see Fig. 2(b)]. In the presence of a temperature change ΔT the heat flow can be expressed as $\dot{Q} = \kappa \Delta T A/\rho$, where κ is the thermal conductivity, A is the area where diffusion occurs, and $\rho = \sqrt{D/f}$ is the thermal penetration depth with D the thermal diffusion and f the frequency ($\rho \approx 100$ μm). Because of the film's thickness, cylindrical diffusion ($A = 2\pi\rho d$, area of a cylinder's wall, where d is the film thickness) is considered while spherical diffusion ($A = 2\pi\rho^2$, area of half a sphere) occurs in the glass substrate. The heat flow ratio obtained is

$$\dot{Q}_{film}/\dot{Q}_{glass} = \frac{\kappa_{film} d}{\kappa_{glass} \rho}, \tag{1}$$

where we have assumed that the temperature change is the same. Equation (1) leads to a heat-flow ratio below 0.13 for Au films less than 40 nm thick on a BK7 glass substrates. In conclusion, as the film thickness increases, the heat flow in the film becomes more relevant and the proportionality hypothesis is no longer valid.

To obtain Γ, we need a sample thick enough that the evanescent wave does not reach the film–air interface but thin enough to be able to disregard the heat flow along the film. Because of the lower thermal conductivity of Pt ($\kappa_{r\_Pt} = 71.6$ W/m·K, $\kappa_{r\_Au} = 317$ W/m·K [10]), we can maintain the heat flow ratio of Au while increasing the film's thickness by a factor of more than 4. Hence, this increase in film thickness, together with the high imaginary part of the dielectric function (15.4 at $\lambda = 539$ nm for Pt, 1.79 at $\lambda = 564$ nm for Au [11]) results in an attenuation ≈ 3000 times greater than in Au films of the evanescent wave. Moreover, with a wavelength of 532 nm the SPP cannot be excited in Pt, since an incidence angle greater than 90° is required. Therefore, scattering losses can be neglected in a Pt film and absorp-

tion can be calculated directly by measuring the reflection at the glass–film interface. In view of the considerations above, we calculate Γ by dividing the measured thermal expansion by the absorbed power, obtaining $\Gamma = \delta L_{z,\mathrm{Pt}}/P_{abs,\mathrm{Pt}}$.

With each thin film, we simultaneously measure the light reflected by the glass–metal interface (Photodiode 2), the thermal expansion by acquiring the FEO signal and fluctuations in the incident power (Photodiode 1) for normalization. To characterize the excitation beam we measure the reflection using a clean glass coverslip. Since we are interested in working only beyond the total reflection angle, this results in a direct measurement of the excitation power, compensating for all the losses from the optical path.

The thermal expansion measurements on the Au films are converted into absorbed power using Γ. Scattering is calculated indirectly by subtracting the reflected and the absorbed from the incident power (transmitted power is null due to total reflection). Experimental results from three different Au thin films (19.5 ± 1.5 nm, 25.5 ± 1.5 nm, and 35.5 ± 1.5 nm) are shown in Fig. 3, where reflection, absorption, and scattering are plotted against the incidence angle of the excitation beam. The rms roughness of each film was measured using a scanning tunneling microscope, obtaining 1.6 nm, 4.6 nm, and 2.7 nm, respectively. For every dielectric function, there is an optimal thickness for which the energy at the film–air interface is maximum, and thus an increase in scattering losses is expected. Our films are in the range of maximum coupling. In Fig. 3(a) the angles for maximum absorption and scattering do not coincide. This

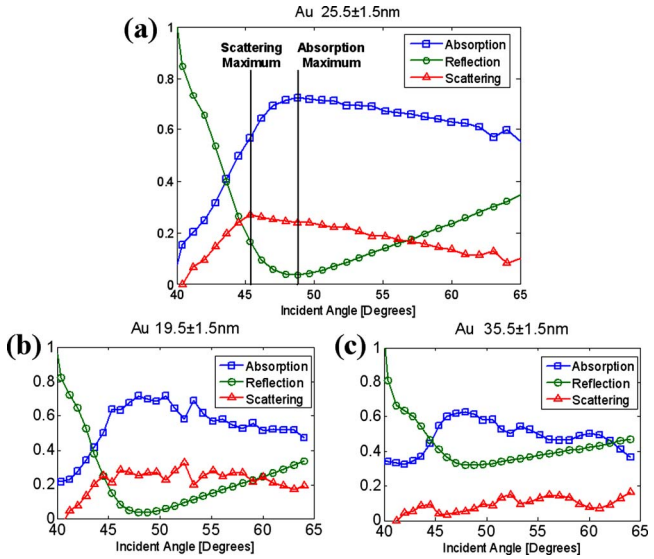

Fig. 3. (Color online) Experimental data for reflection, absorption and scattering on Au films of different thickness: (a) 25.5 ± 1.5 nm, (b) 19.5 ± 1.5 nm, (c) 35.5 ± 1.5 nm. In (a) we can see that the angles of maximum scattering and absorption do not coincide.

angular difference has been observed previously [5].

Through the direct measurement of absorption and reflection, we clearly demonstrate that scattering losses in these films are not negligible at all and show nontrivial angle dependence. Therefore, the next question is how much error one suffers by not considering scattering. For a rough evaluation, experimental reflection and absorption curves were fitted using Fresnel's equations for a simple three-layer model. If scattering is not present, both curves would have essentially the same information (incident power equals the sum of the reflected and absorbed power), and the retrieval parameters should be the same. However, the values for film thickness and complex dielectric function of Au obtained by adjusting both curves differ significantly: 20% of discrepancy in film thickness and 50% in both the real and imaginary part of the complex dielectric function. These results emphasize the need of scattering measurements and new theoretical models that account for interaction of SPP with surface roughness.

In conclusion we show a photothermal technique with high axial ($\approx$10 pm, which corresponds to a temperature incresease of the glass substrate of $\approx$10 mK) and lateral resolution ($\approx$0.5 μm, the size of the diffraction limited spot) capable of characterizing extinction losses, discriminating between absorption and scattering on nanostructures. The thermal expansion is measured using a commercial CD pickup head that possesses a high displacement sensitivity of $\sim$1 pm/μV. Experimental results on thin Au films are presented showing models should include scattering losses for consistent retrieval of optical constants.

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