COMPARISON OF EMISSIVITY MEASUREMENTS USING AN INTEGRATING SPHERE REFLECTOMETER AND A LASER POLARIMETER ON SURFACES WITH VARIOUS DEGREES OF ROUGHNESS

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Abstract – Laser polarimetry has been used for years to obtain normal spectral emissivity measurements on pulse-heated materials [1, 2]. The method is based on the FRESNEL equations that describe reflection and refraction at an ideally smooth interface between two isotropic media. However, polarimetry is frequently used with surfaces that clearly deviate from this ideal condition. Questions arise with respect to the applicability of the simple FRESNEL equations to non-specularly reflecting surfaces. On the other hand, reflectometry utilizing integrating spheres provides a measurement of the hemispherical spectral reflectance for normal incidence, from which the normal spectral emissivity[1] can be derived, regardless of surface texture. In a first effort to explore the limits of polarimetry in terms of surface roughness, room temperature measurements were performed on a number of samples using both an integrating sphere reflectometer and a laser polarimeter. In this paper, the two methods are briefly described and the results of the comparison are discussed.

Keywords: emissivity; integrating sphere; polarimetry; reflectance; reflectometry; rough surfaces; roughness.

1. INTRODUCTION

Shock-physics experiments are routinely performed at the Los Alamos National Laboratory (LANL) as a method to derive the equations of state of materials. In these experiments a sample made of the material of interest is shock-compressed with high explosives (HE) or through the impact of a flyer plate that has been accelerated to a very high velocity. Flyer plates can be laser-driven, HE-driven, or accelerated in a gas-gun. With all these methods extremely high strains and strain-rates can be achieved in the sample.

Temperature is an important parameter in the constitutive equations of materials but its accurate measurement in shock-compression experiments remains a challenge. Because of the very short duration of such experiments (typically a few microseconds), pyrometry is commonly used to obtain shock temperatures.

A measurement of the true (as opposed to radiance) sample temperature requires a pyrometer that is calibrated in terms of blackbody radiance, as well as some knowledge of the normal spectral emissivity of the sample at the operating wavelength of the pyrometer.

Several methods to obtain normal spectral emissivity are known: comparing the thermal radiation emitted from the surface of interest to the thermal radiation emitted from a blackbody cavity at the same temperature [3], integrating sphere reflectometry [4], laser polarimetry [1], or measuring the absorptance ratio at two wavelengths [5], just to mention the more common.

In the shock-physics community, multi-wavelength pyrometry has widely been used to determine the temperature and normal spectral emissivity of the shocked samples [6]. Recently, efforts have been underway to utilize reflectometry (either with or without integrating spheres) [7, 8] and laser polarimetry [9] in these experiments, to dynamically measure emissivity and improve the accuracy of the temperature measurements.

The contactless character of laser polarimetry is a very convenient feature in the face of the highly destructive nature of shock-compression experiments, where any equipment that is placed close to the sample is usually severely damaged. Its weakness lies in the constraints that it places on the sample surface in terms of texture and the presence of oxide layers. The insensitivity of integrating sphere reflectometry to these factors is this technique's greatest advantage.

1.1. Laser polarimetry

Laser polarimetry is based on the measurement of the change in the state of polarization of a laser beam upon reflection at the surface of a sample. Figure 1 shows a schematic of the measurement geometry.

From the measured change in polarization the index of refraction n and extinction coefficient k of the sample can be derived using the FRESNEL equations [10]. These equations describe the reflection of an optical plane wave at the planar interface between two isotropic media in terms of the amplitudes and phases of the incident and reflected waves.

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[1] Strictly speaking, emissivity is an intrinsic material property. The term "emittance" should be used instead, whenever the influence of the surface texture (and possibly surface oxides) on the radiative properties of a sample cannot be neglected. However, this distinction is rarely made in the literature and we will use "emissivity" in all cases in this paper.
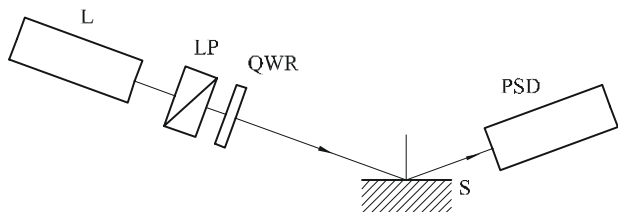
Fig. 1: Schematic drawing of a laser polarimeter. L, laser; LP, linear polarizer; QWR, quarter-wave retarder; S, sample; PSD, polarization state detector.

From the optical constants n and k, the reflectance of the sample for normal incidence is computed. Making use of KIRCHHOFF's law in the energy balance between the incident, absorbed, and reflected light fluxes at the surface of an opaque sample, the normal spectral emissivity of the sample is finally obtained from:

$$\varepsilon = \frac{4n}{(n+1)^2 + k^2} \tag{1}$$

with: ε, normal spectral emissivity; n, index of refraction; k, extinction coefficient. This equation is only valid for vacuum (or air) to metal interfaces.

The validity of the FRESNEL equations, and hence the applicability of laser polarimetry, is limited to ideally smooth surfaces for which reflection is perfectly specular. The difference between reflection from a planar and a rough surface is illustrated in Fig. 2. At a rough surface multiple reflections can occur, which, among other things, will lead to a depolarization of the reflected beam and render the use of the FRESNEL equations impossible.
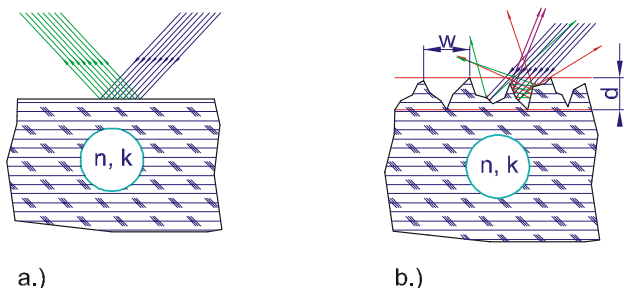

a.)   b.)

Fig. 2: Reflection of a light beam at (a.) a smooth and (b.) a rough surface. The blue rays indicate incident light, green indicates single reflection, and red multiple reflections.

More details about data reduction and the application of laser polarimetry to measurements on pulse-heated samples can be found in Refs. [1] and [2].

1.2. Integrating-sphere reflectometry

As the name implies, this technique employs an integrating sphere to collect laser light that is reflected by the sample into the whole hemisphere above it. Multiple reflections on the highly and diffusely reflecting inner surface of the sphere distribute the light uniformly over the entire sphere in a short time. It is a relative measurement,

where the signal obtained using the sample is referenced to that obtained using a reflectance standard, whose reflectance is accurately known. In the ideal case, the ratio of the radiances produced inside the sphere is equal to the ratio of the reflectances of the sample and the standard. Consequently,

$$\frac{S_s}{S_r} = \frac{R_s}{R_r} \tag{2}$$

with: S_s, signal obtained with the sample; S_r, signal obtained with the reference; R_s, reflectance of the sample; R_r, reflectance of the reference material.

For opaque samples, the normal spectral emissivity is obtained from the measured spectral hemispherical reflectance for normal incidence using KIRCHHOFF's law in the energy balance between the incident, absorbed, and reflected light fluxes at the sample surface:

$$\varepsilon = 1 - R_s \tag{3}$$

with: ε, normal spectral emissivity of the sample; R_s, spectral normal-hemispherical reflectance of the sample.

A schematic drawing of a simple integrating sphere reflectometer is shown in Fig. 3. It can be seen that in order to prevent the specularly reflected component from exiting the sphere after only one reflection, the entrance port is not located directly above the sample, but at a small angle off the normal. This is usually neglected when Eq. 3 is applied.
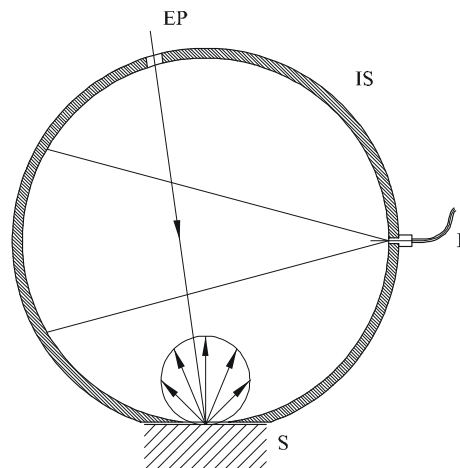

Fig. 3: Schematic diagram of an integrating sphere reflectometer. EP, entrance port; S, sample; IS, integrating sphere; F, fiber to detector.

Figure 3 shows a substitution sphere [11]. Because the average sphere wall reflectance, of which the sample is a part, changes when the sample is substituted for the reference material, the throughput of the sphere is slightly different in the two measurements and an error results from the use of Eq. 2. This error can be avoided by using a more elaborate design. The so-called comparison sphere features an additional sample port that allows the reflectance standard to be mounted on the sphere at the same time as the

sample so that the average sphere wall reflectance is the same in the two measurements.

More details on integrating sphere reflectometry as applied to dynamic emissivity measurements on pulse-heated samples can be found in Ref. [4].

2. MEASUREMENTS

2.1 Instrumentation

A Division-of-Amplitude-Photopolarimeter (DOAP) was used for the polarimetric measurements. It is a commercial instrument that was purchased from Containerless Research (CRI) and has since been upgraded to more closely meet our demanding field requirements. It is a high-speed instrument capable of measuring emissivity every 20 ns. A laser diode that is used in series with an Erbium-Doped-Fiber-Amplifier (EDFA) serves as the light source and can deliver up to 2 W at 1.55 μm. The output power level can be adjusted continuously by varying the current through the laser diode. Some details about this particular instrument can be found in Ref. [9]. More details about DOAPs in general can be found in Refs. [12 - 14].
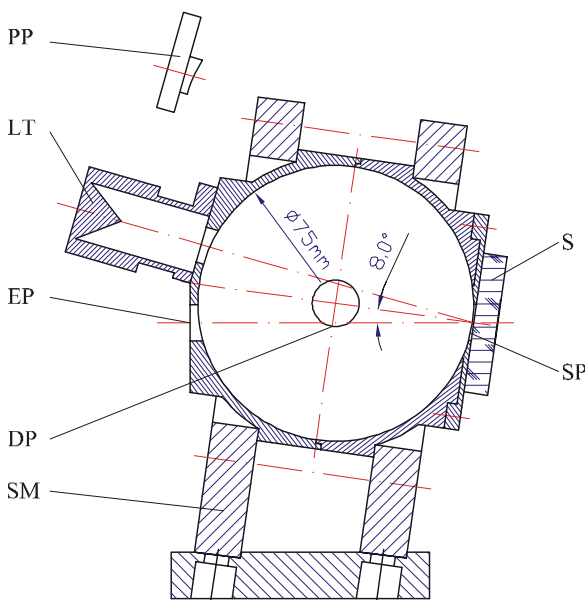

Fig. 4: Schematic of the integrating sphere reflectometer. PP, port plug; LT, light trap; EP, entrance port; DP, detector port; SM, sphere mount; S, sample; SP, sample port.

For the spectral-hemispherical reflectance measurements we built a simple reflectometer with a 70 mm diameter, gold-coated integrating sphere at its heart. The sphere, which was purchased from Gigahertz Optik in Germany, features four circular ports (see Fig. 4) whose total area is less than 5% of the sphere surface area to ensure high sphere efficiency. The same laser that was used with the polarimeter was also used with the reflectometer. A collimated and mechanically chopped laser beam passed through the entrance port and illuminated the sample or the reflectance standard at an angle of 8° off the normal. After being spatially integrated by the sphere, the reflected light was detected by an InGaAs detector that was fiber-coupled to the detector port of the sphere.

It is known that sphere non-uniformity results in a systematic error when the sample and the reference differ in their directional scattering characteristics [15]. This becomes particularly apparent when one tries to measure the reflectance of a highly specular sample using a highly diffuse reflectance standard and vice versa.

In an effort to reduce this kind of error, using a specular and a diffuse reflectance standard, we made two reference measurements for each sample measurement. The spectral hemispherical reflectance of the sample for normal incidence was then computed (in units of per cent) according to Eq. 4 as the weighted sum of the two reflectance values that were measured with the two reference standards.

$$R_s = S_s \cdot \left(\%spec \cdot \frac{R_{s.r.}}{S_{s.r.}} + \left(100 - \%spec\right) \cdot \frac{R_{d.r.}}{S_{d.r.}} \right) \qquad (4)$$

with: S_s, signal measured with the sample; $S_{s.r.}$, signal with the specular reference; $S_{d.r.}$, signal with the diffuse reference; $R_{s.r.}$, reflectance of the specular reference; $R_{d.r.}$, reflectance of the diffuse reference; %spec, percentage of light reflected in the specular direction.

The weighting factors take into account the specularity of the sample surface. In order to determine the value of the quantity %spec in Eq. 4, an additional measurement was made for each sample, this time with the light trap installed on the fourth sphere port instead of the port plug, which normally is part of the sphere surface. The role of the light trap is to subtract (absorb) light reflected by the sample in the specular direction and within the solid angle subtended by this fourth port at the center of the sample. Similarly, a measurement was done with the diffuse reflectance standard and the light trap. Equation 5 was then used to compute %spec:

$$\%spec = 1 - \frac{S_{s,LT}/S_s}{S_{d.r.,LT}/S_{d.r.}} \qquad (5)$$

with: $S_{s,LT}$, signal measured with the sample and the specular light trap; $S_{d.r.,LT}$, signal measured with the diffuse reference and the specular light trap.

Our diffuse standard was purchased from Labsphere® and had a reflectance of 99.2% at 1.55 μm, as stated on the calibration certificate. As a specular standard we used a copper disc whose surface was diamond-turned and whose reflectance of 96.6% at 1.55 μm was computed from the literature values of n and k at that wavelength [16].

2.2 Samples

Measurements were performed on eight samples each of copper, tin, aluminum 1100 (>99.0 weight% Al, 0.12 weight% Cu) and aluminum 6061 (97.9 weight% Al, 0.60 weight% Si, 0.28 weight% Cu, 1.0 weight% Mg, 0.2 weight% Cr). The samples were 40 mm diameter discs with a thickness of 6.4 mm. For each material one sample was polished, one was diamond turned, and six had surfaces

with various degrees of roughness. Of the six rough samples, half were "randomized" and the other half "non-randomized". These two types correspond to what is often referred to in literature as samples with "randomly rough" and "contoured" surfaces, respectively. The randomized samples had their surfaces treated with abrasive paper to make any regular structure disappear, whereas the non-randomized were left with the tooling marks on their surfaces. Since they were fabricated on a lathe, these marks were circular, as illustrated in Fig. 5, and were expected to give rise to a grating effect, which, if true, would definitely be detrimental to polarimetry.
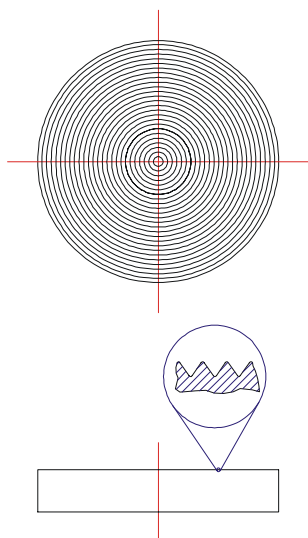

Fig. 5: Conceptual drawing showing the contoured surface (tooling marks) of the non-randomized samples.

The three samples in each group (randomized and non-randomized) had surfaces with an RMS-roughness of 8 µinches, 16 µinches, and 32 µinches. These values were measured using a stylus profilometer (Federal Products Surfanalyzer 5000®).

With each sample, measurements were taken at three locations with the polarimeter and at five locations with the reflectometer. The measurements taken with each method were averaged and a standard deviation was computed. Each polarimeter measurement itself represents the average of 4096 data points.

2.3 Results

Table 1 lists the values of n and k at 1.55 µm, as reported in Ref. [16], for polished copper, tin and aluminum.

Table 2 lists the measured specularity, as expressed by Eq. 5, for all copper samples. The laser power necessary to make these measurements with comparable signals is also shown. These values were normalized with respect to the power used for the measurement on the polished surface. As expected, higher power was required for the rough samples. Similar results, in terms of specularity and laser power, were obtained for all materials.

Table 3 summarizes the reflectance results, as measured with both the polarimeter and the reflectometer, for all samples.

Table 1: The optical constants n and k at 1.55 µm for polished copper, tin and aluminum from Ref. [16]. The reflectance R_{comp} was computed from these values for normal incidence.

material	n	k	R_{comp}(%)
Cu	0.61	8.26	96.55
Sn	3.31	8.67	85.87
Al	1.44	16.0	97.80

Table 2: The measured specularity, according to Eq. 5, of the copper samples, and the (normalized) laser power required to make the reflectometry measurements with comparable signals.

surface type	%spec	normalized laser power
polished	100	1
diamond turned	100	1
8 µinch / randomized	9.5	1.6
16 µinch / randomized	4.2	2.8
32 µinch / randomized	4.1	12.0
8 µinch / non-randomized	56.4	1.2
16 µinch / non-randomized	55.9	1.2
32 µinch / non-randomized	52.2	1.2

The index of refraction n and extinction coefficient k of the various copper samples, as measured with the polarimeter at 1.55 µm, are plotted in Fig. 6.

Figure 7 shows the reflectances of all tin samples, measured by both methods. The error bars shown in this plot indicate the standard deviation of the measurements at different locations on the samples.

Table 3: Reflectance (in %) at 1.55µm for four different materials with various degrees of surface roughness, as measured with the integrating sphere reflectometer (ISR) and the division-of-amplitude-photopolarimeter (DOAP).

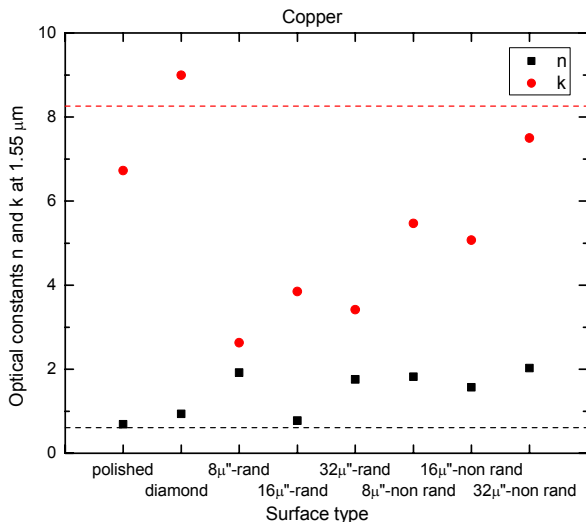| surface type	Cu		Sn		Al 1100		Al 6061	
	ISR	DOAP	ISR	DOAP	ISR	DOAP	ISR	DOAP
polished	97.1	94.42			91.4	93.8	93.4	87.4
diamond turned	96.6	95.76	82.6	85.7	79.4	73.0	91.8	91.1
8 µinch / randomized	69.4	50.4	61.1	66.3	88.2	68.7	78.7	67.4
16 µinch / randomized	87.1	82.8	67.7	58.8	76.6	67.0	86.6	54.0
32 µinch / randomized	82.9	63.7	78.7	74.0	88.5	65.4	84.5	69.6
8 µinch / non-randomized	97.2	80.6	76.5	71.8	93.3	73.2	92.2	87.9
16 µinch / non-randomized	89.2	80.4	76.2	52.0	89.4	76.3	91.3	87.5
32 µinch / non-randomized	94.1	95.1	79.7	82.1	91.7	93.8	88.9	86.7

Fig. 6: The index of refraction n and extinction coefficient k as measured with our polarimeter on the copper samples. The dashed lines represent values from Ref. [16]
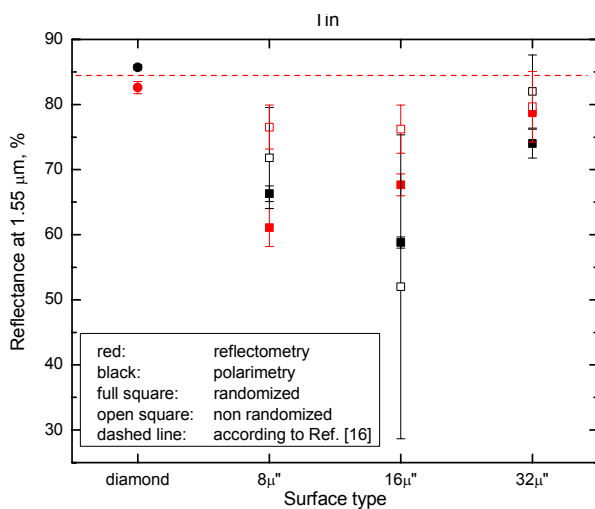

Fig. 7: Measured reflectance of tin on samples with various degrees of roughness. The error bars indicate the standard deviation of the measurements.

3. UNCERTAINTIES

The uncertainty in the polarimetric emissivity measurements on polished and diamond turned samples is believed not to exceed 3% [1]. In the case of measurements on rough samples, it does not make much sense to speak of an uncertainty, when the technique itself might not be applicable. The largest deviation of the polarimetry results from the measurements obtained using the reflectometer was 4% for the randomized samples and 20% for the non-randomized samples.

We have not yet conducted a rigorous uncertainty analysis of the reflectometry measurements. Using the standard deviation of the measurements at different sample locations as a rough estimate, we believe that the uncertainty does not exceed 5% and 8% for the randomized and non-randomized samples, respectively.

4. DISCUSSION

As expected, the measurements obtained with polarimetry and reflectometry on the polished and diamond turned samples generally agreed well with each other and with literature. However, in the case of the aluminum samples the agreement was not that good. We do not understand the reason for that. It is conceivable that the disagreement was caused by the fact that the samples were not of pure aluminum and that the effect of surface oxides was larger in the case of the aluminum samples.

Reflectometry and polarimetry were also in reasonable agreement in the case of the randomized samples, as can be seen in Fig. 7 for tin. Interestingly enough, the polarimetry results followed those obtained with the reflectometer as the surface type changed, even though in this case it is not clear what the physical meaning of the measured n and k values is.

This was not true in the case of the non-randomized samples. The polarimetric measurements deviated significantly from those obtained with the reflectometer. We suspect that this was related to the grating effect that is caused by the periodicity in the roughness of these samples (see Fig. 5). Probably for the same reason, the standard deviation between polarimetric measurements at different locations on the non-randomized samples was significantly larger than in the case of the randomized samples. This is expected, because the polarization of the reflected beam strongly depends on the orientation of the periodic grooves on the sample with respect to the plane of incidence.

More research will be required to really understand our observations. It can already be stated that polarimetric emissivity measurements on anything other than specular surfaces should be treated with caution. It seems to be the case that randomly rough surfaces are more forgiving than contoured surfaces in this respect. Even though reflectometry utilizing integrating spheres is better suited for measurements on rough surfaces, there is a considerable number of potential sources of error relating to the characteristics of the sphere and their deviation from ideal behavior. Using a combination of diffuse and specular reflectance standards can certainly reduce some of these errors to the point where reflectometry becomes a viable option for use in shock experiments.

5. ACKNOWLEDGMENTS

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