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The specular reflection of polarised light from coated paper

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ABSTRACT

An experimental examination of the reflection of polarised light ($\hat{s} \otimes \hat{p}; \lambda = 633$ nm) from surfaces which have a significant specular component is presented. The reflected light intensity is measured using a detector with a narrow acceptance angle (0.5^o) and it is found, for a number of different surfaces based on coated paper, that as the detector scans through the angular distribution, the ratio of polarised light intensities (R_p/R_s) changes in a systematic way which can be accurately predicted if the surface is modelled as an array of mirror-like facets each with the same refractive index, distributed in and around the mean plane of the coating. The change in the ratio R_p/R_s with detector position is independent of the surface.

Key words: specular reflection, polarisation, coatings, paper, surface texture.

INTRODUCTION

The visual appearance of coated paper is of considerable commercial significance with the degree of specular reflection a major factor. Industrial Glossmeters (1,2) are designed to estimate specular reflection at specified angles with specified incident and reflected optical geometries. The reflected intensity is usually compared to that from an optically smooth dielectric surface such as black glass with a specified refractive index. Variable-angle reflection goniophotometers have also been designed and used with the aim of obtaining additional information about the manner in which the reflected intensity varies around a particular specular angle (3,4,5). This angular intensity variation can then provide information about the surface topography of the surface as well as further information relating to the visual appearance (6,7,8). The work described in this paper demonstrates how the reflection of such surfaces. measured polarised liaht from with a reflection goniophotometer, provides new information which a simple model for the surface topography can account for.

EXPERIMENTAL APPARATUS

The reflection goniophotometer shown in Figure 1 was built to measure the reflected intensity of polarised light from small surface areas. The light source is a 2mW He-Ne laser (with polarised output at 633 nm) which is clamped in a fixed position. The polarised beam passes through a rotatable half-wave retarder plate and is then incident on the sample which is held in position on a vertically mounted vacuum-plate which rotates about a central vertical axis; the incident light intercepts this rotation axis and the angle of incidence can be selected by rotating the sample table using a stepper motor. A half-wave retarder plate is used to adjust the plane of polarisation of the incident beam relative to the plane of incidence on the sample. The method of vacuum mounting is particularly convenient for samples in the form of thin sheets provided that the suction holes are remote from the measurement area so that distortion of the sample surface does not occur. The reflected beam intensity is detected by a silicon photodiode which is mounted on the wall of an integrating sphere so that only light depolarised by multiple reflection reaches the photodiode surface. Reflected light from the sample enters the integrating sphere through two in-line circular apertures whose diameters and relative positions from the surface define the acceptance angle of the detector. In the case of the measurements reported here an acceptance angle of 0.5° was used. The detector system (apertures, integrating sphere and photodiode) rotates about the same vertical axis as the sample and both sample and detector angular positions can be selected by means of stepper motors operated from a microcomputer. The angular movement of the detector system can be selected in angular increments from 0.01 degrees with the light intensity being measured 200 times and averaged before moving to the next angular setting. A typical detector scan for the samples described in this work is through 10° in 0.1° steps taking about 30 seconds to complete the scan.

The output from the photodiode is amplified and fed to the A/D converter port of the microcomputer. As only intensity ratios rather than absolute values are required, calibration of the detector is not necessary and any time-varying output from the laser has not been a limiting factor. In the case where accurate recording of the detector output is required, for example when recording the reflected intensity as a function of the state of polarisation of the incident light for subsequent computation, a digital voltmeter is used.

The angular resolution of the system was measured experimentally using polished black glass in place of the vacuum table. With the incident angle set at 65° or 75° , the detector was scanned through the reflected beam in steps of 0.01°. The recorded output is shown in Figure 2. The angular resolution, estimated as the full width at half height, is approximately 0.5° .

Because the incident light is coherent and the reflecting surface is rough, the reflected light contains spacial intensity variations or speckle which are superimposed on the larger scale intensity variations which are the subject of this investigation. However, the speckle is on a sufficiently small angular scale to be effectively integrated over the detector aperture. This was investigated by recording the scale of the speckle pattern intensity variation on a recording medium placed at the position of the first aperture. The recording medium used was a gelatin film containing methylene blue (9). Exposure to the reflected beam from a typical sample of coated paper for about 1 minute caused bleaching of the methylene blue to occur on a scale which is significantly smaller than that of the aperture size. This conclusion agrees with that of other published work (10) in which it was also found that speckle noise was averaged out in the angular cone of measurement.

EXPERIMENTAL PROCEDURE

Specimens were taken from both laboratory and commercial samples of coated paper. In some cases these were also over-printed with a continuous ink film. With a sample mounted on the vacuum plate, the plane of polarisation of the incident light is selected to be in either the s or p mode. With the incident light set at 75° to the sample surface normal, corresponding to the specular angle commonly used for coated paper (1), the detector was programmed to scan through 5° on either side of the specular angle.

A typical result for coated paper is shown in Figure 3 where two scans have been made wth the light incident at 75^o on the same spot on the surface in the s and p modes. The reflected intensities R_p and R_s , for incident polarised light are significantly different. The polarised reflectance ratio R_p/R_s was

also determined at different angular settings of the detector as it passed from 70° to 80° and these are illustrated in Figure 4 where it can be seen that there is a significant increase in the value of (R_p/R_s) as the reflected light is received at increasing angles from the surface normal.

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RESULTS AND DISCUSSION

The angular distribution of reflected light from the coated paper in Figure 3 can be seen to have a number of significant features which are listed below.

- 1. The intensity varies with angle in a non-smooth manner with both the R_{o} and R_{s} distributions having angularly coincident peaks and troughs.
- 2. There is a significant difference between the reflected intensities of the \hat{p} and \hat{s} polarised light.
- 3. The reflected intensities have fallen to insignificant values when the detector position is about 5^o on either side of the specular peak.
- 4. The maximum reflected intensity is not precisely coincident with the specular angle.

The angular coincidence of features in the R_p and R_s distributions is taken to indicate that the detected light originates from precisely the same area of surface in both cases and that rotation of the half-wave retarder plate has not introduced any significant lateral movement of the incident light beam.

The fact that the reflected intensity is very small outside the specular zone indicates that there is no significant contribution to the detected signal from diffusely scattered light originating from within the coating layer at this particular angle of incidence and with this particular detection geometry.

It is to be noted that the large difference between R_p and R_s at the specular angle (75^o) is similar in magnitude to that for a dielectric surface with a refractive index of about 1.5. In addition, the angular variation of the reflected intensity distributions could arise from an array of mirror-like facets which lie in and about the mean plane of the surface with a normal distribution of angles. Such a model has been used in the past (11) to account for the angular distribution of reflected light from rough surfaces. This model is, in many respects, similar to that of a random rough surface discussed by Beckmann (12).

The surface facet model may, however, be extended to account for the polarisation phenomenon illustrated in Figure 4 by noting that whereas those facets lying in the plane of the surface exclusively contribute to the reflected intensity in the specular direction and will therefore have light incident on them at the specular angle, other members of the facet population lying at small angles to the mean plane will thereby have light incident on them at a different angle. In this case it is to be expected that the ratio R_p/R_s would be different to that observed in the specular direction. This can be quantified in

The facet model of the surface is illustrated in Figure 5 where the angle of incidence (relative to the mean plane) is θ . A detector setting of ϕ will then receive light from those facets whose angular setting α is given by

 $\alpha = \frac{\phi \pm \theta}{2}$ (The sign depends on the sense of rotation of the 2 facet relative to the mean plane)

The real angle of incidence on these facets is θ' where

$$\theta' = \theta + \alpha = (\theta \pm \theta/2) + \phi/2.$$
 E1

If the facets each have the same effective refractive index, n, then it is possible to predict how the ratio R_p/R_s should change with ϕ by applying the Fresnel equations, with the appropriate incident angle, to light reflected into the detector at a particular angular setting ϕ

We note that the appropriate refractive index for the surface can be estimated from the ratio of R_p/R_s measured in the specular direction ($\phi = \theta$) by (13,14)

n = sin
$$\theta$$
 [1 + (1- θ) ²/ (1 + θ)². tan² θ]^{1/2} E2
where = (R_p/R_s)^{1/2}

and that this value of n can then be used to predict the value of (R_p/R_s) at a detector setting ϕ by transposing E2 to give

$$(R_p/R_s)\phi t = (1-A)/(1+A)$$
 E3

where A = $[(n/\sin\theta')^2 - 1]^{\frac{1}{2}}/\tan\theta'$

where θ' is given in terms of the detector position ϕ from E1 above.

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APPLICATION

The application of E2 and E3 to the case illustrated in Figure 3 results in the calculated values of the R_p/R_s given in Figure 4 where the comparison with the experimental data shows excellent agreement. It therefore appears that a value of n = 1.446 applies to the majority of the facets in this particular coating.

The value of the refractive index of a coated paper surface has been found to depend on a number of factors such as the type of mineral pigment, the quantity of adhesive used and the degree of compaction produced in the calendering process which is applied after the coating has dried. The effect of changes in the value for n produce consistent changes in the ratio of (R_p/R_s) as illustrated in Figure 6 for different surfaces. Included in Figure 6 are surfaces to which an additional layer of ink or varnish has been applied as this can change the optical properties of the surface significantly. A typical ink film might have a thickness of about a micron and, because of its polymeric and pigmented composition, will have an effective refractive index which can be significantly different to that of the coating layer to which it is Again good agreement is found between experimental and applied. predicted changes in the ratio of R_p/R_s with detector position. Even in the case of a cyan (blue) ink which has a significant optical absorption cross section at the wavelength of the incident light, the rate of change of Rr/Rs with detector position is found to be predictable with a real value for n.

Many rough surfaces have directional properties arising from their method of formation. Because paper is formed from a flowing slurry of pulp fibres picked up on a moving drainage wire, it retains some bi-directional properties which have extreme values in the paper-machine direction and at right angles to it. This has been found to be the case for the reflectance properties measured with the reflectance goniophotometer. An example is shown in Figure 7 where it can be seen that the specular intensity is greater and the intensity distribution is narrower in the machine direction (M) compared with the cross-machine (XM) direction. However, measurement of the ratios R_p/R_s in the two orthogonal directions for the same specimen shows very similar behaviour (Figure 8) because the refractive index in the two directions are very similar. This emphasises again that it is the refractive index which controls the value of R_p/R_s at a particular detector setting rather than the distribution of facet angles which compose the surface.

During the preparation of coated paper surfaces for examination by Scanning Electron Microscopy it is necessary to apply a thin conducting coating by vapour deposition to prevent electrostatic charging. When the conducting layer applied is gold, the optical properties of the surface are changed significantly and will be characterised by a complex refractive index with a significant absorption contribution. As the values of n and k for a metal surface will depend upon the method of deposition it was decided to first determine these properties from a layer of gold formed on a smooth plastic substrate under similar conditions of vapour deposition to those used subsequently on the coated paper samples.

In contrast to the determination of n from E2, where only the ratio R_p/R_s is required, it is necessary to have individual values of both R_p and R_s in order to determine n and k. This was done by using a dielectric surface of known refractive index to calibrate the detector in terms of the absolute values of R_p and R_s given by the Fresnel equations.

Measured values of R_p and R_s for a vapour deposited gold surface on a flat plastic substrate are shown in Figure 9. The values of n and k were determined from the equations given below, derived from the standard Fresnel equations for a metal surface with conducting electrons.

$$n = \frac{(1/\cos^2 \theta - \cos^2 \theta)/2}{[(R_p + 1)/\cos \theta (1 - R_p) - (R_s + 1)\cos\theta/(1 - R_s)]}$$

k = [2n (R_p + 1) / cos θ (1 - R_p -) - n² - cos⁻² θ]^{1/2}

Values of n=0.256 and k=2.126 were obtained in this way for this particular gold film. (For comparison, data for electropolished gold (15) interpolated to a wavelength of 0.633 μ m gives n=0.118 and k=3.26. The difference is assumed to arise from the methods of surface formation).

A similar application of gold to a commercially printed surface was found to give a ratio of (R_p/R_s) which was essentially constant over the detector settings from 70° to 80° when the incident angle was 75° (Figure 10). This is in contrast to the changing value of R_p/R_s found for the dielectric surfaces at this incident angle studied above. Using the estimated values of n and k from the smooth gold surface, calculations were made for the ratio R_p/R_s over the angular range of interest with the results also shown in Figure 10. It can be seen that the calculated ratio R_p/R_s is nearly stationary although its value is slightly greater than the experimental value. This is taken as confirmatory evidence for the facet model of the surface.

DISCUSSION

Neither shadowing nor multiple scattering between adjacent facets, which would result in cross-polarisation, seems to occur to any appreciable extent on the surfaces examined. The facets have been assumed to have definable Fresnel reflection coefficients associated with them so any micro-roughness on the facet surface is small enough to give essentially specular reflection at the high angles of incidence used in the present work. The requirement for specular reflection may also impose a lower limit on the area of a facet if diffraction and scatter are not to dominate.

The types of surface studied here have topographical features on at least two scales; one corresponds to the relatively long correlation lengths associated with the underlying substrate structure which is composed of pulp fibres, whilst the other is on a much smaller scale associated with the dimensions of the pigment particles used in the coating. This type of surface has been described as a composite roughness model (16) and equations have been derived which describe the reflection intensity to be expected for any combination of incident or reflected angle, state of polarisation, surface slope, dielectric constant and small scale roughness. The considerable computation involved with these equations precludes their ready use to confirm the above experimental observations and it is suggested therefore that the simple facet model still has some utility.

Note that the change in the ratio ${\sf R}_p/{\sf R}_s$ with changing angle of view for a dielectric surface under the conditions reported here depends only on the value of n and is independent of the actual distribution of facet angles in the surface.

It is noted that Stagg and Charalampopoulos (17) have shown that the determination of the refractive index of a rough surface from polarised angular reflectance values can be made with reasonable accuracy over a range of specular angles. We have used one incident angle only (75^o), chosen to correspond to the specular angle used industrially. It is fortuitous that this is very close to the optimum incident angle of 74^o proposed by Miller (18) for determining optical constants.

CONCLUSIONS

A reflectance goniophotometer has been built which can measure the angular distribution of reflected polarised light from optically rough surfaces. Coated and printed paper surfaces which have a significant specular reflection at high incident angles have a distribution of reflected intensity extending to a few degrees on either side of the specular peak which shows a physically significant change in intensity when the polarisation state of the incident light is changed from being parallel (R_p) to perpendicular (R_s) to the incident plane.

The ratio R_p/R_s increases numerically as the angle of the received light is increased. This increase can be accurately predicted from a value of the effective refractive index of the surface together with a simple model of the surface topography based on small specularly reflecting areas of the surface (facets) which lie in and about the mean plane of the surface.

Applying a vapour deposited gold layer to the surface produces a change in the ratio R_p/R_s with position in the reflected light which is consistent with a complex refractive index for the surface.

It is believed that the utility of these observations lies in giving credibility to the facet model which allows a simple practical interpretation of the optical properties of a general class of rough surfaces to which coated and printed paper belong.

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FIGURE 2 Response of reflection goniophotometer using polished black glass: (incident angle 65^o and 75^o.



FIGURE 3 The reflected intensity from coated paper as a function of detector angle. The incident light is polarised in either the s or p mode.



FIGURE 4 The polarised reflectance ratio R_p/R_s as a function of detector position for the coated paper sample illustrated in Figure 3.



Schematic cross section through a coated surface with reflecting facets



Deflection from specular angle is $(\Phi - \Theta) = 2\alpha$

FIGURE 5 The facet model of a rough surface.



FIGURE 6 A comparison of measured and calculated values for the polarised reflectance ratio (R_p/R_s) for coated surfaces with a range of refractive index values. Data points are indicated by symbols; full lines are calculated values.



FIGURE 7 The reflected intensity distribution from coated paper relative to the direction of sheet formation. M = machine direction; XM = cross-machine direction.



FIGURE 8 The polarised reflectance ratio R_p/R_s values as a function of detector position for the data illustrated in Figure 7. Symbols indicate experimental data points; full lines are calculated.



FIGURE 9 Polarised reflectance from a vapour deposited film of gold on a smooth plastic substrate. The R.H. axis refers to the measured intensities given in detector output voltages. The L.H. axis refers to the ratios R_p/R_s both measured from the curves and calculated from n=0.256 k=2.126.



FIGURE 10 Similar data to that given in Figure 9 but for gold vapour deposited on a coated paper surface.

THE SPECULAR REFLECTION OF POLARISED LIGHT FROM COATED PAPER

L F Gate & D J Parsons

Additional contribution:

Measurement of Void Volume in Calendered Coatings by Specular Reflection

It was demonstrated previously(1) that it is possible to derive a value for the refractive index (n) of a coated or printed surface from measurements of the specular reflection using polarised incident light. In this supplement the interpretation of the numerical value of n found for paper coating is considered in more detail.

A paper coating is composed of mineral particles glued together with adhesive and contains a significant number of sub-micron sized air voids which can account for up to 30% of the total volume of a calendered coating as determined by mercury porosimetry. These three major components can each be characterised by its own refractive index which will contribute to the value of the bulk coating refractive index. In order to relate the bulk refractive index of the coating layer to the constituent components we note that n appears to be linearly dependent on the volume fraction of each phase (2,3,4) Thus in the case of suspensions of latex and kaolin, measurements of n determined by critical angle reflection showed a linear relationship to the solid phase up to volume fractions of 0.5 in the case of latex and 0.1 for kaolin. (Investigations into higher fractions were precluded by the method of measurement used.) In the case of porous coatings, the voids occupy a smaller fractional volume than the solid phase so that the voids may be regarded as the dispersed phase in the coatings.

We may use this evidence for a linear relationship as a model for the coating layer and write its refractive index as:

 $n = n_s \bullet V_s + n_v \bullet V_v$ E1

where n_s = refractive index of solid phase V_s = Volume fraction of solid phase

 $n_v = refractive index of void (= 1)$ $V_v = volume fraction of voids (=1-V_s)$

where
$$V_s + V_v = 1$$

Hence $V_v = (n_s - n)/(n_s - 1)$ E2

In order to obtain a value for n_s we may apply the additive rule to the components making up the solid phase so that we write:

E3

 $n_s = n_p V_p + n_a \bullet V_a$ where n_p = pigment refractive index

 V_p = volume fraction of pigment

 $n_a = adhesive refractive index$

V_a = volume fraction of adhesive

where $V_p + V_a = 1$

In the case of a typical offset coating formulation using kaolin (density = 2.64 g/cm^3) and latex (= 1.09 g/cm^3) with 10 pph latex we

find $V_a = 0.21$ and $V_p = 0.79$. A value of $n_p = 1.57$ can be used for the kaolin at the He-Ne laser wavelength of 633nm while $n_a = 1.59$ (5) in E3 then gives $n_s = 1.574$.

To illustrate the use of E2 to calculate the void volume, a series of offset coatings based on latex (10pph) and kaolin were applied to a base paper and calendered at 4 increasing pressure settings using 10 nips each.

The ratios of R_p/R_s were measured at 75° and used to calculate a value for the refractive index (n) of the coating surface at each pressure (1). These values of n were then used in E2 to calculate a value for the coating void volume at each calender pressure with the results shown in Table 1.

Calender	Rp/Rs at 75°	Refractive index	Fractional void in
Pressure Kg/cm	incidence		surface layer
5	0.299	1.431	0.250
10	0.293	1.444	0.228
14.4	0.283	1.465	0.191
20	0.275	1.483	0.160

Table 1: Effect of calender pressure on coating structure.

Note that with increasing calender pressure, the refractive index of the coating increases significantly; this translates, with the aid of E2, into a decreasing void volume in the surface layer. Figure 1 illustrates that the decrease in void volume is linearly related to the calender pressure applied.



Figure 1: Effect of calender pressure on fractional void in surface layer.

It should be noted that the above estimates of void volumes made by surface reflection must necessarily be a measure of the values within about 0.5 μ m of the interface with air. In the case of uncalendered coatings applied to a non-porous base there is good agreement between the void volumes measured by mercury porosimetry and the specular reflection technique. Calendering the coating results in a lower estimate of void volume by reflectance suggesting that the surface layer may be more compressed than the bulk. Recent published measurements using an image analysis technique applied to electron micrographs of coatings (6) have also indicated that coatings can be denser at the surface.

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Dr D Priest, UMIST, UK

In your curves of specular reflectance against angle there almost always appear to be distinct peaks and valleys. What are the significance of those?

L Gate

We are looking at a surface area of about 3mm x 1mm and from the facet model, I think we are seeing, non uniform angular distributions of facets which are giving us this rather irregular appearance to the spectral curve. If we were looking at a very much larger area, I think that the peaks and valleys would be very much smoother; what we see is evidence for the areas of the coating which are distorted by the fibrous structure underneath.

Prof B Lyne, Royal Institute of Technology, Sweden

Can you say what the size of the examination spot is – the reason why I am asking is I wonder if you can look at the variation locally in the refractive index and deduce local variation in the concentration of binder in the coating which is very important for mottling?

L Gate

At the moment we are limited to an area of about 3mm x 1mm when the setting is at 75° incident angle. This is determined by the incident laser beam diameter hitting the surface and the angular resolution of the detector. To go to smaller areas it would be necessary to focus the beam on the surface. We would then lose the very low divergence of the laser beam which might be a disadvantage. Yes, it would be a sensible way to progress the investigation miaht and vou beain to pick up smaller inhomogeneities in the surface.

B Lyne

Do you in fact see variation in the refractive index when you scan across a sheet of paper?

L Gate

There is a variation. It tends to be smaller as you increase the calendering.

Prof D Wahren, Stora Teknik AB, Sweden

Congratulations – a beautiful paper. I have a problem trying to find some of the references you mentioned in the text. Could we please have the additional material you have presented – including the very

interesting illustrations?

L Gate

Yes, that is in the hands of the Chairman for inclusion in Volume 3. (See additional text at the beginning of the discussion of the paper.)

I Kartovaara, Enso-Gutzeit Oy, Finland

Can you tell us what is the measurement volume or measurement depth from the surface of the coating?

L Gate

Bearing in mind we are using laser light at 633 nm, it is probably of the order of half a micron. It is determined by the depth of the evanescent wave. We have also to remember that we are looking at a slightly rough surface. I would guess that it is about half a micron. The interesting thing is that if we put down an ink film which is only about a micron thick we get quite a sensible answer for the refractive index. The depth is certainly less than a micron.

Dr D Page, Pulp & Paper Research Institute, Canada

Following up on that point I am rather puzzled that you have added in the volume of voids to calculate the refraction index. Is each individual void very small, less than a wavelength of light, because it would not work if it were not.

L Gate

Yes. The theory behind the simple mean value formula is extremely complex and I don't think there is a firm idea of what the upper limit to pore size would be. I can only refer to the references for latex particles up to 1 micron in size; in a coating. The void volume is composed of pore radii of about 0.1 micron.

Prof H Kropholler, UMIST, UK

Do you think the results of the variability that you have got could be related to print mottle?

L Gate

Yes. However I think that it is a very big subject for which we have not yet got the experimental evidence.