



# LUND UNIVERSITY

## Time-resolved Studies of Light-propagation In Paper

Carlsson, J; Hellentin, P; Malmqvist, L; Persson, Anders; Persson, W; Wahlström, Claes-Göran

*Published in:*  
Applied Optics

*DOI:*  
[10.1364/AO.34.001528](https://doi.org/10.1364/AO.34.001528)

1995

[Link to publication](#)

*Citation for published version (APA):*

Carlsson, J., Hellentin, P., Malmqvist, L., Persson, A., Persson, W., & Wahlström, C.-G. (1995). Time-resolved Studies of Light-propagation In Paper. *Applied Optics*, 34(9), 1528-1535. <https://doi.org/10.1364/AO.34.001528>

*Total number of authors:*  
6

### General rights

Unless other specific re-use rights are stated the following general rights apply:

Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal

Read more about Creative commons licenses: <https://creativecommons.org/licenses/>

### Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

LUND UNIVERSITY

PO Box 117  
221 00 Lund  
+46 46-222 00 00

# Time-resolved studies of light propagation in paper

J. Carlsson, P. Hellentin, L. Malmqvist, A. Persson, W. Persson, and C-G. Wahlström

A method for time-resolved recording of light scattering in thin, highly scattering media is described. Subpicosecond pulses from a high-power Ti:sapphire laser are used, and single-shot recordings of the scattered light are made with a fast streak camera. The method is applied to the study of light scattering in paper, and a 1-ps resolution is demonstrated. The dependence of the light scattering on the basis of weight and density of the paper has been studied. A white-light continuum generated from the high-power pulses by the use of self phase modulation in water is used to study the wavelength dependence of the scattering process. A model for the propagation of light in paper has been developed and used in Monte Carlo simulations. The experimental results are used for testing this model, and absorption and scattering parameters are determined from that comparison.

*Key words:* Paper, light propagation, light scattering, time-resolved spectroscopy, white-light continuum, streak camera, Monte Carlo simulations.

## 1. Introduction

The way that light propagates in noncoated paper, newsprint in particular, affects the possibility of printing high-resolution pictures on such paper. Present models for light scattering in paper are mostly based on the Kubelka–Munk theory.<sup>1–3</sup> To test these models optical properties such as reflection and transmission are often used. To obtain a more detailed understanding of light propagation in paper, experiments that show in better detail what happens to the light are needed. One possibility for doing this is to measure with a high time resolution the delay of a light pulse passing through a sheet of paper. Time-resolved light scattering has previously been used to determine scattering parameters, for example, for a suspension of polystyrene latex spheres in a 4-cm-long cell.<sup>4</sup> In such thick samples delays of hundreds of picoseconds occur because of scattering.

If a short pulse of light is directed onto a sheet of paper and the temporal shape of the pulse after it passes the sheet is recorded, properties such as the

variation of the path inside the paper may be determined. Conversely, if a light-scattering model is used to simulate the passage of light through a sheet of paper, the change of the shape of a light pulse will depend in a detailed way on the absorption and scattering cross sections, the angular distribution of the scattering, the distance between the scattering centers, and the number of scattering events. The experiments will therefore be a critical test of any model for the propagation of light in paper. The speed of light is 0.3 mm/ps. Considering the thickness of a sheet of paper, it is obvious that the time resolution needed for such experiments is of the order of 1 ps. Because of the increasing use of color printing, these experiments need to be performed for different wavelengths in the visible region. Such measurements are presented in this paper. The method that is used is applicable to the study of any thin, highly scattering medium.

## 2. Experimental Method

The light source used in this study is a high-power laser<sup>5</sup> that is able to produce pulses with an energy per pulse of more than 200 mJ and a peak power of more than 1 TW. The high-power pulses are produced by taking single pulses from a mode-locked continuous Ti:sapphire laser and amplifying them repeatedly in Nd:YAG-pumped Ti:sapphire amplifiers. The pulse-repetition rate is 10 Hz. The pulses have a duration of 0.2 ps. The laser is tunable between 760 and 840 nm but was used at the fixed wavelength

---

J. Carlsson, A. Persson, W. Persson, and C-G. Wahlström are with the Department of Physics, Lund Institute of Technology, P.O. Box 118, S-221 00 Lund, Sweden. P. Hellentin and L. Malmqvist are with the Swedish Newsprint Research Centre, Danderydsvägen 95, S-182 65 Djursholm, Sweden.

Received 14 December 1993; revised manuscript received 8 September 1994.

0003-6935/95/091528-08\$06.00/0.

© 1995 Optical Society of America.

of 796 nm. For the present application the most interesting property of this light source is the very short duration of the pulses. The pulses are shorter in space than the thickness of the studied paper sheets. Only a very small part of the available pulse energy is used.

To perform time-resolved studies of light scattering at any wavelength in the visible region, a white-light continuum<sup>6</sup> is generated by focusing the high-power pulses in water. As a result of the Kerr effect the refractive index of a medium changes in proportion to the intensity of the incident light. This causes a frequency shift of the light that is proportional to the time derivative of its intensity. This self phase modulation will thus be most prominent in the case of an incident pulse with high peak intensity and short rise and fall times.

For the generation of the white-light continuum the 796-nm light is focused in a cuvette that contains water. A pulse energy of 15 mJ is used. The white light generated in the water is collected with a second lens and used for the experiment. This procedure and the setup are illustrated in Fig. 1. To study light scattering at a particular wavelength, the white light can be filtered out from the continuum. In this study this was done with various interference filters, each with a 10-nm bandwidth. The beam had a diameter of 0.5 mm at the surface of the paper.

Experiments were also performed at the fundamental wavelength of the laser, at 796 nm, and at 398 nm. The 398-nm light was produced by frequency doubling the laser light in a 2-mm KDP crystal.

The light transmitted through the paper sample was detected with a fast streak camera (Hamamatsu Model C 1587). This was used to detect a single transient of light transmitted through the paper as one of the subpicosecond pulses was focused on the paper. Single-shot detection has the advantage of yielding the best possible time resolution. It does, however, yield signals with rather poor dynamics because the intensity of the detected light has to be very low. A higher intensity of the light would yield a higher density of photoelectrons in the streak tube, and the repulsion between the electrons would cause the resolution to deteriorate. Signal averaging could improve the dynamics, but at the expense of the time resolution.<sup>7</sup> In a single-shot experiment the response function of the detection system, that is, a recording of the laser pulse, has a full-width at half-maximum (FWHM) of typically 2.5 ps. In the present studies a resolution of 1 ps is desirable, making single-shot experiments the more favorable

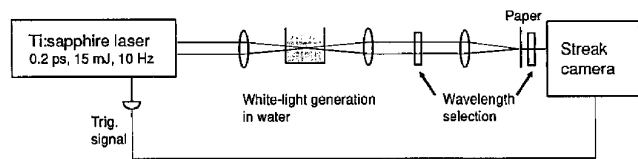


Fig. 1. Experimental setup for time-resolved measurements of light scattering in a single sheet of paper.

technique. To avoid any influence from fluorescence, the detection is made through an interference filter that transmits only the wavelength of the incident light.

Figure 2 shows a recording of a single pulse at 796 nm scattered through one sheet of newsprint and also a direct recording of the laser pulse. The pulse width is increased by 2 ps because of the passage through the paper. The thickness of the sheet is 70  $\mu\text{m}$ . Cellulose has a refractive index of close to 1.5, which means that if light had passed straight through the sheet the transit time would have been 0.35 ps. It should be pointed out that the increase of the width of the recorded curve measures not the transit time but the variation in transit time between different photons. If all photons had the same transit time, the width of the pulse would not be affected. In the example shown in Fig. 2 the variation in transit time is approximately 2 ps.

### 3. Paper Samples

To facilitate studies of the dependence of light scattering on individual properties of the paper, paper sheets with well-controlled properties were prepared. Three series of sheets were manufactured. Within each series only one property was varied. The paper samples are described in Table 1. The first series was made from pure thermomechanical pulp (TMP), and the sheets were made with different basis weights but approximately the same density. Papers with weights from 30 to 153  $\text{g}/\text{m}^2$  were made. The second series was made from the same pure TMP, but in this series the weight was kept as close to 80  $\text{g}/\text{m}^2$  as possible, while the density was varied by the use of different wet pressings (yielding densities of 300 and 400  $\text{kg}/\text{m}^3$ ) and by calendering of the 400  $\text{kg}/\text{m}^3$  paper with different pressures, yielding papers with higher densities. Densities of between 300 and 723  $\text{kg}/\text{m}^3$  were thus obtained. The third series was made from 6-mm rayon fibers, yielding a highly porous structure. These sheets were made with weights from 50 to 90  $\text{g}/\text{m}^2$ . Finally, measurements were also made on ordinary newsprint.

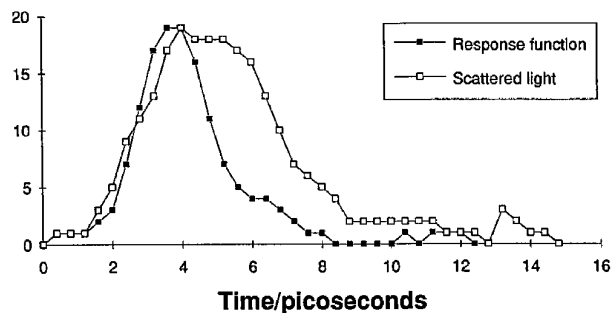


Fig. 2. Time-resolved recording of a 0.15-ps pulse of light at 796 nm scattered through one sheet of newsprint. Response function is a recording of the laser pulse. Scattered light is a recording of the pulse that has passed the 70- $\mu\text{m}$ -thick sheet of newsprint.

Table 1. Properties of the Studied Papers

Material	Basis Weight (g/m <sup>2</sup> )	Thickness (μm)	Density (kg/m <sup>3</sup> )	Reflection		Transmission	
				500 nm	700 nm	500 nm	700 nm
TMP	30	90	330	0.44	0.44	0.29	0.33
	49	135	365	0.53	0.59	0.14	0.22
	70	182	383	0.55	0.65	0.077	0.16
	90	225	402	0.56	0.69	0.047	0.12
	111	270	411	0.56	0.72	0.021	0.078
	133	314	422	0.56	0.73	0.007	0.063
	153	357	428	0.56	0.75	0.002	0.045
	80	270	300	0.56	0.67	0.052	0.14
	80	200	400	0.56	0.67	0.054	0.14
	84	146	575	0.55	0.67	0.045	0.14
	79	132	597	0.55	0.67	0.056	0.14
	81	134	608	0.56	0.67	0.055	0.14
	81	125	649	0.56	0.67	0.056	0.14
	79	114	698	0.56	0.67	0.050	0.14
82	113	723	0.55	0.67	0.054	0.14	
Rayon fiber	50	~1000	~50	0.40	0.44	0.36	0.38
	70	~1400	~50	0.57	0.52	0.30	0.28
	90	~1800	~50	0.61	0.55	0.26	0.26
Newsprint	45	70	640	0.47	0.54	0.18	0.24

4. Measurements

The transmission of the sheets and the sensitivity of the streak camera varies with the wavelength. To maintain the recorded intensity constant, independent of the wavelength, the single-shot recordings were performed with pulse energies between 0.01 and 6 μJ. These pulse energies correspond to between  $2.0 \times 10^{10}$  and  $1.2 \times 10^{13}$  photons per pulse. We varied the pulse energy by entering different neutral density filters in the laser beam. The response function was recorded with much lower pulse energy.

Two recordings in each of three points were made for each sheet and each wavelength. The response function was recorded before, in between, and after the recordings with paper, in 15 occasions at all for each wavelength.

Figure 3 shows a direct recording of the laser pulse together with four recordings of a laser pulse that has passed through a sheet of paper. The four recordings were made with sheets that have different basis

weights. Figure 4 shows similar recordings for sheets with different densities but the same basis weight. Both Figs. 3 and 4 show recordings made at 796 nm. Figure 5 shows recordings of pulses at three different wavelengths that have passed a sheet of paper with 111-g/m<sup>2</sup> basis weight.

In addition to the time-resolved measurements, the absolute reflection and transmission of the sheets have been determined. This was done with an integrating sphere, which measured either the total reflected intensity or the total transmitted intensity when light of a selected wavelength was focused on a sheet of paper. The light source used was a high-pressure xenon lamp. The illuminated area was a circle with an 8-mm diameter, large enough to average out local variations in the paper. For two of the studied wavelengths, 500 nm and 700 nm, the results are included in Table 1. Figures 6 and 7 illustrate

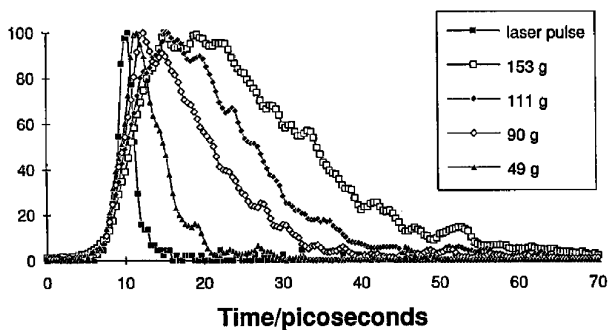


Fig. 3. Response function of the detection system (2.5-ps FWHM) and single-shot recordings of light pulses at 796 nm that have passed single sheets of paper with different basis weights. Weights are given per square meter.

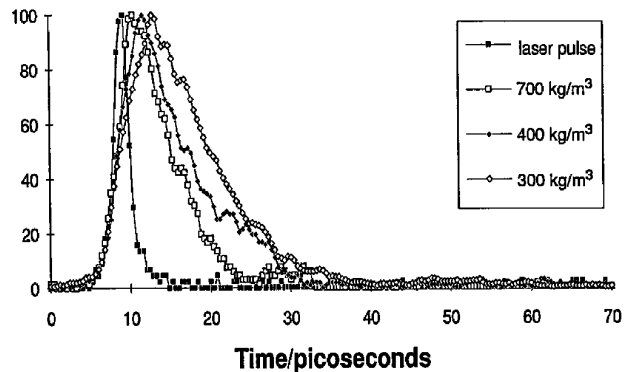


Fig. 4. Response function of the detection system (2.5-ps FWHM) and single-shot recordings of light pulses at 796 nm that have passed single sheets of paper with the same basis weight but different densities.

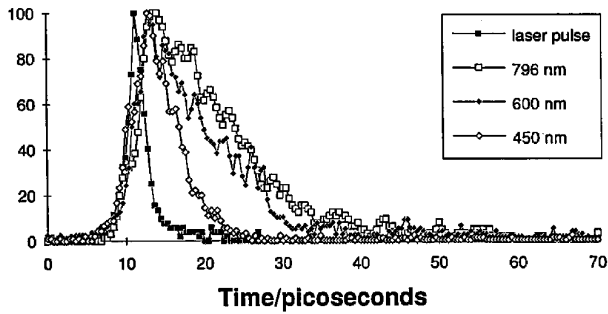


Fig. 5. Time-resolved recordings of sub-picosecond pulses of different wavelengths that have passed a single sheet of paper with an 111-g/m<sup>2</sup> basis weight.

the wavelength dependencies of the reflection and transmission for some different sheets.

### 5. Experimental Results

For each recording of the time distribution of the scattered light the FWHM, and the positions of the maximum and the center of gravity have been evaluated. The results are summarized in Tables 2–4. The values given are the average and, in parentheses, the standard deviation for the 6 or 15 recordings. The standard deviations of the evaluated parameters for the laser pulse and for the light that is scattered through the sheets with the lowest basis weights are typically 0.5 ps. This means that although the width of the response function is as large as 2.5 ps, changes of less than 1 ps can be determined.

The absolute time scale is not accurate enough for comparison of different recordings on a subpicosecond level. In the determination of the FWHM or the time distance from the peak to the center of gravity this is of no consequence. However, when determining the shift of the center of gravity caused by the scattering in the paper, one would need a fixed time scale for comparison with the recordings of the laser pulse. For such comparisons an internal calibration has been used: for each curve the time at which it reaches 20% of its peak value has been used as the starting point for the time scale.

Tables 2–4 show three different parameters for the recorded curves: the width of the curve, which yields the variation in transit time; the time from the peak of the curve to its center of gravity, which is a

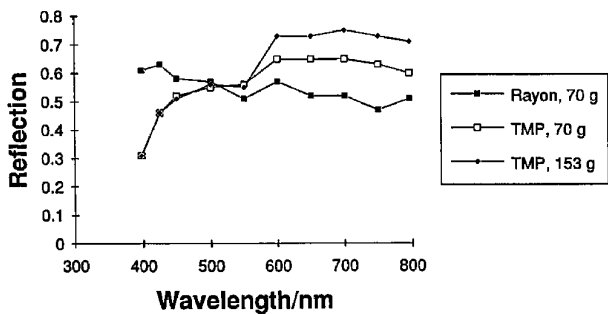


Fig. 6. Absolute reflection at different wavelengths for three different paper sheets.

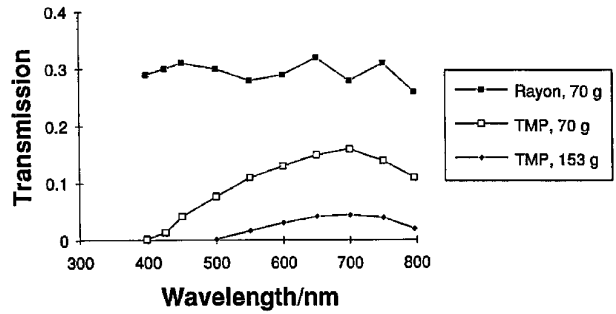


Fig. 7. Absolute transmission at different wavelengths for three different paper sheets.

measure of the asymmetry of the transmitted pulse; and the shift of the center of gravity compared with the response function, which is the average extra time spent in the paper by the transmitted photons because of the scattering. Because there is no absolute time scale for comparison of different recordings, the third parameter becomes more uncertain: these values may be slightly too small.

For the TMP sheets with different basis weights the values of all three parameters increase when the basis weight is increased. This is to be expected because light will take longer to go through a thicker paper. The effect is, however, very different at different wavelengths. In Fig. 8 the dependence of the shift of the center of gravity on the basis weight is illustrated for different wavelengths. At 398 nm the increase is hardly significant. This means that only very few of the transmitted photons have been scattered along a long path inside the paper. This could be due to either a small probability of scattering at short wavelengths or a large probability of absorption. For the papers with the highest basis weights no measurements could be performed at 398 nm because of the low transmission.

The shift increases with the wavelength to reach a maximum at 700 nm. At the longest wavelength, 796 nm, the shift again decreases. As can be seen in Fig. 7, the transmission also has a maximum at 700 nm for sheets made from TMP.

The paper with the highest basis weight has a thickness of 0.35 mm. With a refractive index of 1.5 it would take 1.8 ps for light to travel straight through this sheet. At 700 nm the shift of the center of gravity is 9 times larger, indicating that the average path of red light that passes through this sheet is 10 times the thickness of the paper.

For the sheets with different densities but the same basis weight, all three parameters decrease with increasing density. The reason for this may be that a higher density means a shorter distance between the scattering centers in the paper and thus a short time of passage. Again the effect is most notable at the longer wavelengths.

For the three sheets made of rayon fibers the scattering parameters increase as the basis weight is increased. In this case the increase is also seen at the shortest wavelengths. One difference between the rayon and the TMP sheets is that the rayon

**Table 2. FWHM's of the Recorded Curves (in Picoseconds) for Different Wavelengths**

Paper Sheet	398 nm	450 nm	500 nm	600 nm	700 nm	796 nm
Laser pulse	2.5 (0.4)	2.8 (0.5)	2.7 (0.5)	2.3 (0.5)	2.6 (0.4)	2.5 (0.4)
TMP						
Basis weight (g/m <sup>2</sup> )						
30	2.8 (0.3)	3.4 (0.4)	3.9 (0.4)	3.1 (1.0)	3.8 (0.5)	3.0 (0.4)
49	2.4 (0.4)	3.6 (0.4)	5.0 (0.5)	2.8 (0.9)	4.7 (0.2)	3.7 (0.8)
70	3.1 (0.5)	4.8 (1.0)	5.7 (0.6)	5.3 (1.2)	7.0 (0.4)	4.3 (1.1)
90	3.1 (0.6)	5.3 (1.2)	7.2 (1.2)	5.9 (1.8)	7.5 (1.8)	8.0 (1.4)
111	3.2 (0.5)	6.6 (1.2)	9.2 (1.0)	8.8 (0.6)	10.9 (3.0)	13.6 (1.4)
133		7.6 (0.9)	11.0 (0.7)	11.1 (2.4)	15.3 (1.6)	12.7 (1.7)
153		5.4 (1.8)	12.1 (1.2)	10.7 (2.5)	16.1 (3.0)	18.6 (1.4)
Density (kg/m <sup>3</sup> )						
300	3.0 (0.6)	5.9 (0.1)	6.3 (0.8)	6.0 (1.5)	8.4 (1.8)	9.3 (1.2)
400	3.2 (1.0)	5.1 (0.8)	5.2 (0.9)	7.7 (1.9)	7.5 (1.5)	8.4 (1.2)
575	1.9 (0.3)	4.7 (1.4)	4.8 (0.5)	6.3 (1.8)	7.2 (1.0)	8.1 (1.3)
597	2.8 (0.4)	4.8 (0.5)	5.1 (0.6)	5.8 (1.1)	7.4 (0.8)	6.9 (1.6)
608	2.5 (0.4)	5.3 (0.8)	5.8 (1.3)	6.4 (0.9)	7.3 (1.3)	6.7 (1.1)
649	2.5 (0.7)	4.6 (0.4)	5.4 (1.3)	6.8 (1.0)	6.4 (0.9)	6.2 (0.2)
698	2.9 (0.2)	4.5 (0.6)	5.4 (2.1)	5.9 (0.8)	6.6 (0.9)	6.2 (0.7)
723	2.6 (0.3)	4.4 (0.8)	5.0 (0.6)	6.1 (1.4)	8.0 (1.3)	6.5 (1.3)
Rayon fiber						
Basis weight (g/m <sup>2</sup> )						
50	3.2 (0.7)	3.9 (0.4)	3.4 (0.5)	2.6 (0.6)	3.6 (0.7)	5.8 (0.5)
70	3.5 (1.3)	3.8 (0.5)	3.8 (0.3)	3.7 (0.9)	4.0 (0.4)	5.8 (0.8)
90	5.5 (2.1)	4.0 (1.2)	4.2 (1.6)	5.4 (2.7)	6.0 (1.6)	6.9 (1.5)
Newsprint	2.4 (0.4)	4.0 (0.7)	3.7 (0.7)	3.7 (0.6)	3.7 (0.8)	3.1 (0.3)

sheets have approximately the same transmission at all wavelengths in the visible region, whereas the sheets made from TMP have much lower transmission at short wavelengths. It may therefore be

concluded that the reason why the increase in the transmitted pulse length for TMP sheets is almost negligible at short wavelengths is a large absorption rather than a small scattering. At 398 nm, photons

**Table 3. Time from Peak to Center of Gravity of the Recorded Curves (in Picoseconds) for Different Wavelengths**

Paper Sheet	398 nm	450 nm	500 nm	600 nm	700 nm	796 nm
Laser pulse	0.1 (0.3)	0.3 (0.4)	0.4 (0.4)	0.4 (0.5)	0.6 (0.7)	0.2 (0.5)
TMP						
Basis weight (g/m <sup>2</sup> )						
30	0.2 (0.4)	1.0 (0.3)	0.9 (0.7)	0.1 (0.4)	0.2 (0.2)	0.1 (0.7)
49	0.2 (0.4)	0.5 (0.6)	1.7 (0.7)	0.6 (0.5)	1.1 (1.0)	1.4 (0.5)
70	0.0 (0.5)	0.8 (0.7)	2.0 (0.5)	1.2 (1.3)	2.7 (1.4)	2.1 (0.8)
90	0.3 (0.3)	1.3 (0.9)	2.2 (0.5)	1.7 (0.9)	4.3 (1.5)	2.7 (1.0)
111	0.3 (0.3)	1.2 (1.1)	3.0 (1.0)	3.5 (1.6)	6.5 (2.2)	6.5 (0.6)
133		2.2 (1.1)	3.4 (1.8)	5.8 (2.0)	10.4 (1.7)	5.2 (2.3)
153		1.6 (1.4)	2.7 (0.8)	6.2 (3.0)	12.0 (2.3)	6.3 (2.7)
Density (kg/m <sup>3</sup> )						
300	0.4 (0.2)	1.1 (1.0)	1.9 (1.5)	2.9 (1.3)	3.2 (2.1)	3.9 (0.9)
400	0.1 (0.6)	1.0 (1.0)	2.3 (1.1)	3.5 (0.9)	4.6 (1.7)	2.6 (0.7)
575	0.2 (0.3)	1.2 (0.6)	1.6 (1.2)	2.1 (1.0)	2.2 (1.5)	2.6 (0.4)
597	0.2 (0.4)	1.0 (0.7)	1.3 (0.7)	1.6 (0.8)	2.6 (0.8)	3.3 (0.5)
608	0.2 (0.2)	0.9 (0.9)	1.9 (0.6)	2.5 (0.6)	3.6 (0.7)	2.7 (1.0)
649	0.2 (0.4)	0.8 (0.4)	1.0 (0.9)	1.7 (1.3)	3.8 (1.9)	2.6 (0.8)
698	0.1 (0.3)	0.7 (0.7)	2.0 (0.7)	1.3 (1.2)	2.0 (0.6)	2.3 (0.8)
723	0.2 (0.4)	0.8 (0.7)	0.7 (0.4)	1.2 (1.1)	1.9 (1.2)	2.5 (0.9)
Rayon fiber						
Basis weight (g/m <sup>2</sup> )						
50	1.4 (0.7)	1.0 (0.6)	0.7 (0.7)	0.4 (0.5)	0.6 (0.7)	3.2 (0.4)
70	1.7 (0.7)	1.1 (0.6)	3.1 (1.7)	2.7 (0.6)	2.5 (0.9)	3.2 (0.4)
90	4.0 (1.1)	2.0 (0.6)	4.8 (0.4)	5.7 (0.8)	5.8 (1.0)	4.6 (1.4)
Newsprint	-0.2 (0.2)	0.8 (0.4)	0.8 (0.6)	1.0 (0.7)	0.9 (0.2)	0.6 (0.8)

**Table 4. Shift of Center of Gravity of the Recorded Curves Compared with the Recordings of the Laser Pulse (in Picoseconds) for Different Wavelengths**

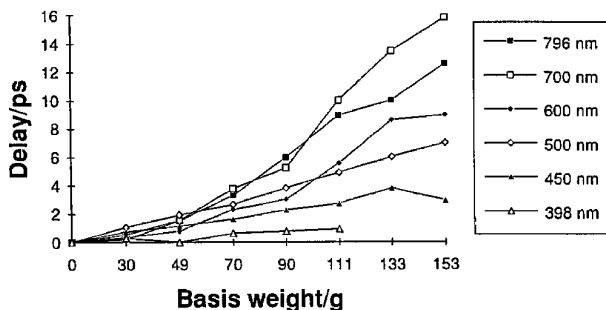
Paper Sheet	398 nm	450 nm	500 nm	600 nm	700 nm	796 nm
<b>TMP</b>						
Basis weight (g/m <sup>2</sup> )						
30	0.3	0.8	1.1	0.4	0.6	0.2
49	0.0	1.2	1.9	0.8	1.6	1.5
70	0.6	1.6	2.7	2.3	3.8	3.3
90	0.8	2.3	3.8	3.0	5.3	6.0
111	0.9	2.8	4.9	5.6	10.0	8.9
133		3.8	6.0	8.6	13.5	10.0
153		3.0	7.0	9.0	15.8	12.6
Density (kg/m <sup>3</sup> )						
300	0.4	2.5	3.6	4.3	4.8	6.0
400	0.5	2.0	3.3	4.5	5.7	5.4
575	0.3	1.6	2.2	3.2	3.8	4.9
597	0.5	1.9	2.2	2.8	3.6	4.8
608	0.2	2.1	2.7	3.1	4.8	4.4
649	0.3	1.8	2.5	3.1	4.7	4.1
698	0.4	1.7	2.5	3.0	3.2	4.2
723	0.3	1.8	2.6	3.2	3.7	4.2
<b>Rayon fiber</b>						
Basis weight (g/m <sup>2</sup> )						
50	1.4	1.2	0.4	0.2	0.7	4.7
70	2.1	1.7	2.6	2.6	2.3	4.8
90	4.3	1.9	4.7	5.9	6.3	6.0
<b>Newsprint</b>	-0.1	0.8	1.0	1.3	0.9	0.3

that are scattered over a long path inside a TMP sheet have a much larger probability of being absorbed than photons at longer wavelengths, and only a few such photons are therefore transmitted.

### 5. Light-Propagation Model and Monte Carlo Simulations

Paper is a network of cellulose fibers. Most of the fibers are aligned in the plane of the paper. Furthermore, most of the originally cylindrical fibers are collapsed and flat.

In the model presented here, paper is taken as a set of parallel cellulose multilayers in air, and light scattering is assumed to occur only in the interfaces of air and cellulose, with refractive indices 1.00 and 1.53, respectively. The interfaces are assumed to be



**Fig. 8.** Shift of the center of gravity of a light pulse that has passed through one sheet of TMP paper as a function of the basis weight of the paper.

parallel to the plane of the paper. Given the refractive indices, the probabilities for reflection and transmission at an interface are calculated with the Fresnel formulas. In the case of transmission the angular change of direction is calculated with Snell's law. The deviation from this simple model of geometric optics is introduced by a random change of direction that occurs at each scattering event, be it reflection or transmission. The word "scattering" is used here for what is really a combination of geometric reflection at the surfaces and a random scattering. The deflection angle of the random scattering is assumed to follow a distribution originally proposed for galactic scattering<sup>8</sup>:

$$p(\cos \theta) = \frac{1 - g^2}{2(1 + g^2 - 2g \cos \theta)^{3/2}},$$

where  $g$  is the expectation value of  $\cos \theta$  and  $\theta$  is the deflection angle. The adjustable parameter is the anisotropy  $g$ . It can vary between  $-1$  and  $1$ , where  $1$  is only forward scattering,  $-1$  is only backward scattering, and  $0$  is isotropic scattering. At each scattering event the probability for reflection and transmission was first calculated, and a new direction of propagation was calculated. From that direction a random scattering was made, with the distribution given above. In these simulations this random scattering is only allowed into a half-sphere around the direction first given by the Fresnel and Snell formulas. The azimuthal scattering angle has an isotropic distribution over the interval  $0$  to  $2\pi$ . The distance between interfaces is assumed to have an exponential distribution that is the same when the light is traveling in cellulose and in air.

The probability of absorption is assumed to be the same in all the paper. This approximately is justified when the mean free path for absorption is much larger than the mean free path for scattering. This yields an exponential distribution for the mean free path for absorption.

Using this model for the propagation of light, one can describe paper with three parameters: the mean free path for absorption, the mean distance between scattering interfaces, and the anisotropy. These three parameters may of course depend on the wavelength of the light and may be different for different sheets of paper.

The model for the propagation of light in paper outlined above has been used in Monte Carlo simulations. In the simulations light entered perpendicularly onto the paper, as it did in the experiments. Simulations have been performed with values of the mean free path for absorption of between  $0.1$  and  $50$  times the thickness of the paper, for mean distances between interfaces of between  $0.002$  and  $0.2$  times the thickness of the paper and for the anisotropy between  $0$  and  $1$ . In each simulation the reflection, the transmission, and the average optical path in the paper for transmitted photons have been calculated. The number of different combinations of parameters was  $14,036$ . In each run  $10,000$  photons were used.

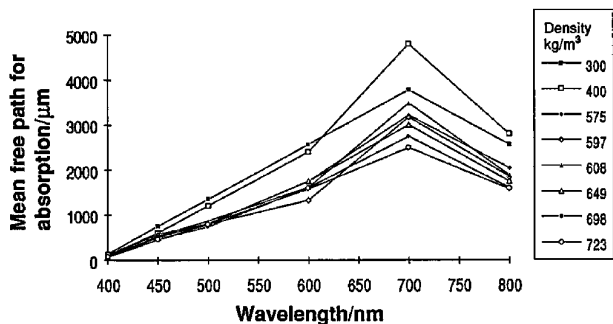


Fig. 9. Mean free path for absorption at different wavelengths for sheets of paper with different density. The values have been obtained by comparison of the experimental results with Monte Carlo simulations.

The experimental data have then been compared with the results of the simulations, and the combination of parameters that best describes each sheet at each wavelength was determined. The experimental value for the average optical path was calculated as the shift of the center of gravity of the pulse times the velocity of light in free space plus the thickness of the sheet.

Of the three series of sheets, the one with the same basis weight but varying density turns out to be the most interesting one. The results for this series are given in Figs. 9–11. Figure 9 shows the mean free path for absorption as a function of the wavelength of the light for the different sheets. There is a strong dependence of mean free path of absorption on wavelength, with a maximum mean free path at 700 nm. The mean free path is roughly inversely proportional to the density, which is very reasonable considering that the change of density has come about by compression of the material. Figure 10 shows the mean distance between scattering surfaces. There is no significant wavelength dependence, but the mean distance is inversely proportional to the density, in the same way and by the same reason as the mean free path for absorption. Figure 11 shows the anisotropy of the scattering. The anisotropy shows very little systematic variation and is typically 0.97 or 0.98. A value of 0.97 for  $\cos \theta$  corresponds to a scattering angle of  $14^\circ$ .

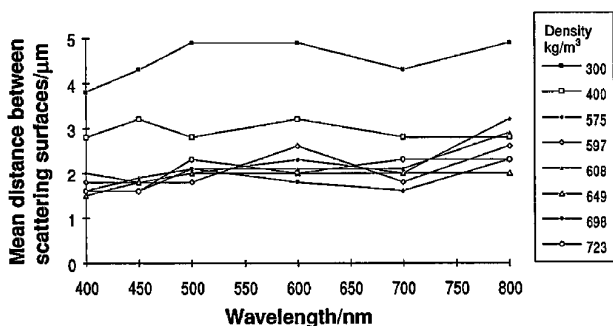


Fig. 10. Mean distance between scattering interfaces at different wavelengths for sheets of paper with different density. The values have been obtained by comparison of the experimental results with Monte Carlo simulations.

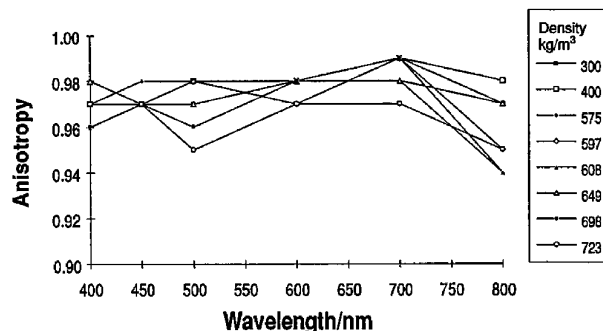


Fig. 11. Anisotropy parameter of the light scattering at different wavelengths for sheets of paper with different density. The values have been obtained by comparison of the experimental results with Monte Carlo simulations.

For the series of sheets with varying basis weight but nearly the same density, most of the sheets show results similar to those of the  $400\text{-kg/m}^3$  sheet in the previous series. This may be expected because the sheets are all produced from the same material. The exceptions are the two sheets with the lowest basis weights, which yield much smaller values for the mean free path for absorption (by factors of 2 or 3). The measurement of the transit time through these thin sheets may be more uncertain than for the thicker sheets.

The third series, made from rayon fibers, does not have the layered structure assumed in the present model and has therefore not been evaluated.

## 6. Conclusion

A technique for a direct time-resolved study of light propagation in paper has been described, and it is found that with a picosecond resolution it is possible to study in detail the passage of light through a single sheet of paper. Furthermore, the dependence of light propagation on the basis weight and density of the paper has been studied. For paper made from TMP the effect of scattering has been found to be strongly wavelength dependent. Sheets made from pure cellulose, on the other hand, do not show any significant wavelength dependence for light within the visible region.

These time-resolved measurements are complemented by absolute measurements of reflection and transmission.

To gain a better understanding of the interaction between light and paper and to understand the behavior of light in a paper structure the experimental results are used in combination with Monte Carlo simulations. A model for the propagation of light in paper has been presented. This model, when used in the Monte Carlo simulations, yields consistent results for the absorption and scattering parameters of systematic series of paper sheets. These calculations also show that it is indeed the large variation in the mean free path for absorption that causes the wavelength dependence.

The interaction of light with the paper structure is one of the most important properties of graphic



paper. One objective in the production of paper for graphic use is to minimize the transmission and to maximize the reflection of light in the visible region. These time-resolved measurements on well-defined paper structures open up possibilities of a more detailed interpretation of the mechanisms of light propagation in paper. Furthermore, with an improved model of light-paper interaction, optical measurements will yield better possibilities for characterization of paper structure. Such improved methods for characterizing the inner structure of paper are needed to design optimal printing properties of paper in the future.

The authors are grateful to Lars Wågberg, SCA Research, Sundsvall, for providing the systematic series of paper samples and to Annika Nilsson, Department of Physics, Lund Institute of Technology, for her assistance with the reflection and transmission measurements. This work was supported by the Knut and Alice Wallenberg Foundation, the Swedish Council for Planning and Coordination of Research, the Swedish Research Council for Engineering Sciences, and the Swedish Natural Science Research Council.

## References

1. P. Kubelka and F. Munk, "Ein Beitrag zur Optik der Farbanstriche," *Z. Tech. Phys.* **12**, 593–601 (1931).
2. P. Kubelka, "New contributions to the optics of intensely light-scattering materials. Part 1," *J. Opt. Soc. Am.* **38**, 448–457 (1948).
3. P. Kubelka, "New contributions to the optics of intensely light-scattering materials. Part 2: nonhomogeneous layers," *J. Opt. Soc. Am.* **44**, 330–335 (1954).
4. G. Zaccanti, P. Brusaglioni, A. Ismaelli, L. Carraressi, M. Gurioli, and Q. Wei, "Transmission of a pulsed thin light beam through thick turbid media: experimental results," *Appl. Opt.* **31**, 2141–2147 (1992).
5. S. Svanberg, J. Larsson, A. Persson, and C.-G. Wahlström, "Lund high-power laser facility—systems and first results," *Phys. Scr.* **49**, 187–197 (1994).
6. R. L. Fork, C. V. Shank, C. Hirlimann, R. Yen, and W. J. Tomlinson, "Femtosecond white-light continuum pulses," *Opt. Lett.* **8**, 1–3 (1983).
7. S. Andersson-Engels, R. Berg, A. Persson, and S. Svanberg, "Multispectral tissue characterization with time-resolved detection of diffusely scattered white light," *Opt. Lett.* **18**, 1697–1699 (1993).
8. L. G. Henyey and J. L. Greenstein, "Diffuse radiation in the galaxy," *Astrophys. J.* **93**, 70–83 (1941).