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Near–infrared incoherent coupling and photorefractive response time of ‘blue’ Rh:BaTiO_3

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Abstract

We have investigated the time constants of writing and decay of photorefractive gratings in ‘blue’ Rh:BaTiO_3 at visible and near-infrared wavelengths. High reflectivity ($>$ 200 %) double phase conjugation has been achieved at near-infrared wavelengths (730 – 809 nm) and efficient double colour pumped oscillation has been demonstrated between wavelengths separated by 150 nm.

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1 Introduction

Rhodium doped $BaTiO_3$ has interesting nonlinear properties [1]–[4]. This blue-coloured $BaTiO_3$ has a strong absorption in the visible (4.2 cm^{-1} at 647 nm) [3] and in the near-infrared (1.4 cm^{-1} at 800 nm) and an infrared response that extends beyond 1 μm. The enhanced absorption in the infrared opens up the possibility of applications for devices pumped by laser diodes, for example at wavelengths of 800 nm and 980 nm.

Studies of blue $Rh{:}BaTiO_3$ have showed high phase-conjugate reflectivities [3] and large two beam coupling single-pass gains [5] both in the visible and near-infrared. However, to be considered useful in the infrared, a photorefractive material must demonstrate not only a reasonable efficiency but also an acceptable response time. Here we show that using the double phase conjugate mirror (DPCM) configuration reflectivities as high as 200 % and transmissions of 26 % are achievable using near-infrared wavelengths of 760–800 nm. Also, we show the speed of grating formation and decay at visible and near-infrared wavelengths is usable, namely having a value of 10 seconds at moderate intensities (above 125 mW/cm^2). We chose to measure the time response using a coherent beam geometry to write a simple grating, rather than with incoherent beams (as in the DPCM geometry), because the analysis of the results is far simpler and easier to compare with other similar time-constant measurements.

2 Efficient two–beam coupling and its response time

In order to investigate the temporal response of $Rh{:}BaTiO_3$ we measured the build-up times of two-beam coupling gain at two wavelengths, 647 nm and 800 nm. Gratings were written using a constant intensity ratio, $r = I_{pump}/I_{signal}$ equal to 16 at 647 nm and 24.3 at 800 nm. We monitored the temporal build-up of gratings using a weak He-Ne probe beam (300 $\mu W/cm^2$ for the 647 nm case, and 90 $\mu W/cm^2$ for the 800 nm case). Figure 1 is a log-log plot of the writing time versus total laser intensity. A linear fit to both sets of data (shown as solid lines) shows the response time τ depending on intensity as I^{-x}, where x is not equal to 1. Having values of $x < 1$ is usually associated with the presence of additional traps [6, 7]. Similar experiments which we performed in our sample of nominally undoped $BaTiO_3$ yielded the value of $x = 1.01$ [8], consistent with the single species photorefractive model. The sublinearity of time response is consistent with the detailed experimental studies of photorefractive species in $Rh:BaTiO_3$ we performed earlier, where additional secondary centres were suggested to account for strong intensity–dependent change in absorption [9, 10].

Blue $Rh:BaTiO_3$ is relatively fast, approximately three times faster in the visible than nominally undoped $BaTiO_3$ for the same incident intensity. In the infrared, $Rh:BaTiO_3$ responds with strong coupling ($\Gamma = 17$ cm^{-1}) which can build-up on a scale of 100 seconds. This is a significant improvement, as compared with a typical undoped $BaTiO_3$, which exhibits only negligible beam coupling and phase conjugation at such wavelengths. All in all, these new experimental findings can be of interest for applications where an improvement both in speed and sensitivity is critical.

Finally, to complete our response time measurements we investigated both the dark decay, and light-induced grating erasure time of gratings. We measured diffraction efficiency from a two–beam coupling grating probed by a very weak He–Ne, after switching off both writing beams. In the dark, the gratings in the undoped $BaTiO_3$ were washed out extremely slowly, at a rate of 0.12 % per minute. In the $Rh:BaTiO_3$, about 17 % of diffraction efficiency decayed rapidly within the first few seconds. This fast drop in grating diffraction efficiency was then followed by a long (hours) slow decay. The fast decay can be associated with the decay of intensity–dependent absorption effect and thermal excitation from secondary traps present in $Rh:BaTiO_3$. The long term decay usually comes from the decay of deep level gratings [11].

The light–induced erasure times of gratings were measured from the steady–state level of two–beam coupling in the undoped $BaTiO_3$ and from the 'quasi steady–state' level (to which the crystal relaxes following the first rapid decay) in the $Rh:BaTiO_3$. A white light source was used to erase the gratings and the rate of their erasure was measured for different intensities of the white light source.

As can be seen from figure 2 photorefractive gratings decayed more slowly in $Rh:BaTiO_3$ than in undoped $BaTiO_3$. The same inclination of the decay times of both crystals suggests that similar deep traps are responsible for creating gratings. These results confirmed our previous work on characterisation of traps in $Rh:BaTiO_3$ [10].

The time constants measured in two-beam coupling gave us an important indication about the time scale of writing gratings both in the visible and near-infrared. It can be regarded as a certain fundamental limit of photorefractive response of Rh:BaTiO$_3$, which is one of the most essential parameters of a photorefractive material. The other important feature of Rh:BaTiO$_3$ we investigated is its infrared sensitivity, in particular, its ability to couple beams of mutually incoherent infrared light.

3 Double phase conjugation and double colour pumped oscillations in Rh:BaTiO$_3$

A double phase conjugate mirror (DPCM) [12, 13] couples mutually incoherent light beams. When two light beams of identical (DPCM) wavelengths are incident on the opposite faces of a photorefractive crystal, they create a common grating which is then responsible for diffracting each beam into a phase conjugate reflection of the other beam. The incident beams can even have different wavelengths (a scheme often called double colour pumped oscillator: DCPO), but the same mechanism of a shared grating will diffract light into two new beams. DPCM is ideally suited for application in reconfigurable, self-aligning and dynamic optical interconnects and image colour conversion.

Recently theories have been developed to describe the temporal and spatial behaviour of DPCM [14] and its threshold conditions. Also, interesting experimental investigations led to observation of diffraction efficiencies (7.4 %) in the DPCM arrangement at 1.54 μm in CdTe:V [15] and double colour pumped oscillations (985 nm to 1.05 μm) with conversion efficiencies of up to 63 % [16].

In our work we concentrated on determining the reflectivity and transmission of DPCM at several near-infrared wavelengths, compatible with laser diode radiation, namely 730 – 810 nm. Also, we aimed at improving the phase conjugate conversion efficiency by using visible light (647 nm) in one of the beams. So far, the possibility of obtaining incoherent coupling in Rh:BaTiO$_3$ has been demonstrated only in the bird-wing configuration [17] and the results obtained showed only a limited phase–conjugate transmission.

3.1 Experimental arrangement

Figure 3 presents a schematic diagram of the experimental set–up. Two incident beams, originating from an Ar^+ pumped Ti:Sapphire laser, overlapped in a 4.7 × 2.7 × 4.4 mm^3 sample of Rh:$BaTiO_3$. We measured the phase conjugate reflectivity, R_{pc}, as a ratio of the intensity of the phase conjugate beam I_3 to the incident beam I_1. The change in reflectivity was then monitored for different intensities of incident beams, namely $r = I_2/I_1$. Rh:$BaTiO_3$ absorbs light very strongly well into the far end of the visible spectrum (α =2.4 cm^{-1} at 736 nm) and this condition imposed certain limits on the choice of the experimental geometry. As a result, large crossing angles (165 – 175 °), typically used for generating double phase conjugation in undoped $BaTiO_3$ to increase the interaction length, didn't lead to the optimum reflectivity. Instead we used a smaller angle of of 150°. We tested the ability of Rh:$BaTiO_3$ to produce DPCM in the infrared by tuning the Ti:Sapphire laser to wavelengths of 760, 800, 809 and 830 nm.

3.2 Experimental results and discussion

Figure 4 presents the DPCM reflectivity results obtained at the far end of the visible spectrum, i.e. at 736 nm ($R_{pc} = I_3/I_1$). The maximum reflectivity obtained was 75 % for a beam ratio value r up to 25. Figure 5 presents results obtained at 760 nm. In this case we obtained a very promising reflectivity of 205 % over a wider range of intensity ratios (up to 70). Wavelengths further into the infrared didn't yield any higher reflectivities, and at 800 nm we achieved only 83 % (figure 6) and 70 % at 809 nm. Any attempts to produce stable DPCM signals at 830 nm failed due to the strong competition from SPPC.

The double-phase conjugation process in blue Rh:$BaTiO_3$ suffers from a competition from self–pumped phase conjugation (SPPC) occurring for one or both beams. Self–pumped phase conjugation is very prominent in Rh:$BaTiO_3$ and can be generated for a wide range of angles of incidence [3]. It also affects strongly the range of input intensities used for DPCM (see figures 4 – 6). The stable operation of DPCM requires two relatively strong input beams, which limits the range of input intensity ratios. If one of the beams is made too weak (below 3 mW for I_1 with $I_2 = 70mW$ for 736 nm) then DPCM signals get unstable and instead SPPC can build–up. Additionally, the maximum reflectivity obtained in the DPCM measurements may not persist at a steady–state value, but can exhibit a similar decay in time as two–beam coupling gain does in the presence of strong fanning. The problem of achieving a stable phase conjugate reflectivity in blue Rh:BaTiO$_3$ was already encountered in the SPPC configuration [3]. We aim to address this problem in the DPCM case in further experimental studies.

The promising, high reflectivity results for double phase conjugation we present here are, to our knowledge, the first such report in blue Rh:BaTiO$_3$. The results obtained indicate that the highest reflectivity (205 %) can be achieved at a wavelength experiencing an intermediate absorption (1.9 cm^{-1} at 760 nm). At other wavelengths, which encounter either stronger absorption (2.4 cm^{-1} at 736 nm) or weaker absorption (1.4 cm^{-1} at 800 nm and 1.3 cm^{-1} at 809 nm), the DPCM showed much lower reflectivities. The efficiency of the DPCM link in Rh:BaTiO$_3$ is also influenced by several other effects, such as the magnitude of light–induced transparency, strength of fanning and self–pumped phase conjugation at a particular wavelength.

Having high reflectivity DPCM can be particularly relevant and important in applications where efficient phase conjugation is sought for a high quality, but weak, input signal beam which can be pumped in the DPCM configuration by, for example, a powerful but multi–mode input beam. Also, it can be of interest for the DPCM schemes where a link is set–up between light from a single–mode and a multi–mode fibre [18].

Phase conjugate reflectivity, however, is just one factor indicating the efficiency of the DPCM process. The other important consideration is phase conjugate transmission ($T_{pc} = I_3/I_2$). In their work, MacCormack *et al.* [17] showed that the phase conjugate transmission achieved in the bird-wing configuration in blue Rh:BaTiO$_3$ was only 6 %, which is a typical value expected for this particular phase conjugate geometry. Figure 7 shows the maximum transmission we measured in our experiment for four different wavelengths. A relatively high transmission (up to 26 %) was observed, but existed only for input intensity ratios close to 1.

In order to investigate whether the reflectivity of the DPCM can be improved we modified our experimental set-up and changed the wavelength of just one of the input beams. We used a Kr^+ laser (647 nm) as beam 1, and 730 (and then 800 nm) for beam 2. For both arrangements we found that the external crossing angle of 146 ° was the optimum for observation of double colour pumped oscillations (DCPO). The intensity of the oscillation beam I_3 at 730 nm (800 nm) was measured for different input beam intensities and the reflectivity was calculated as a ratio of I_3/I_1. For the 647 nm and 730 nm DCPO, we measured very low reflectivities, reaching only up to 60 %. This would indicate again the destructive effect of strong absorption, affecting now both wavelengths, and that of fanning, prominent with illumination with light from the red region of spectrum. On the other hand for the pair of beams of 647 nm and 800 nm we obtained a significant improvement. From figure 6 it can be seen that in the case when both beams had wavelengths of 800 nm, only a modest 83 % phase conjugate reflectivity could be observed. Figure 8 presents the results of the DCPO with 647 nm and 800 nm, and shows that pumping Rh:$BaTiO_3$ with strongly absorbing 647 nm beam helped to increase the intensity of the oscillation beam I_3 at 800 nm up to 200 %, an improvement by a factor of 2.5. Additionally, the range of input beam intensity ratios increased significantly from the maximum $r = 10$ (figure 6) to $r = 450$ (figure 8). These results prove that high reflectivities of phase conjugate (oscillation beams in DCPO) can be achieved in Rh:$BaTiO_3$, and possibly with the aid of a theoretical model to help with the choice of wavelengths and geometries, higher values of reflectivities can be obtained. This is the first demonstration of efficient coupling via DCPO arrangement between visible and infrared wavelengths, separated by as much as 150 nm.

Transmission and reflection from a DPCM grating can also be calculated theoretically. Analytical expressions for these two magnitudes can be derived [12] for the case of undepleted pump approximation and with negligible absorption. Since we measured earlier the coupling coefficient for 800 nm (which is one of the essential parameters required) [8] we estimated the maximum DPCM transmission and reflectivity. The values obtained, namely 83 % for the maximum predicted transmission, and 500% for reflectivity, are more than three, and six, respectively, times higher than the values observed in our experiment. Such a big difference between experimental and theoretical values clearly indicates that absorption cannot be ignored in this case. Absorption in blue Rh:$BaTiO_3$ is not only strong [3], but also intensity dependent [9, 10]. We are currently working on a numerical model which would include absorption and its change with light intensity and simulate the intensity of phase conjugate beams in double phase conjugation.

4 Conclusions

We have measured the photorefractive response time of blue Rh:BaTiO$_3$ in the visible and near–infrared and determined the speed of writing gratings in this crystal. At its maximum absorption (647 nm), Rh:BaTiO$_3$ responds not only three times faster than a nominally undoped BaTiO$_3$, but the gratings created lead to a extremely efficient beam coupling. Strong and relatively fast response of Rh:BaTiO$_3$ extends also well into the infrared.

We have showed that high reflectivities (205 %) and relatively high transmission (26 %) can be achieved from the DPCM gratings formed at near–infrared wavelengths. However, to achieve more progress we have to address the problem of competition with self–pumped phase conjugation by careful theoretical modelling of the DPCM process and other effects present in Rh:BaTiO$_3$.

In order to test and utilise the full potential of DPCM in Rh:BaTiO$_3$ we modified the experimental arrangement and used a visible (647 nm) beam as one of the input beams. The reflectivity of an infrared oscillation beam was improved via this DCPO arrangement from 83% to 200 %. Unique properties of blue BaTiO$_3$ allowed us to observe and measure such high efficiency power conversion from the visible to the infrared, in spite of a large difference in wavelengths.

5 Acknowledgements

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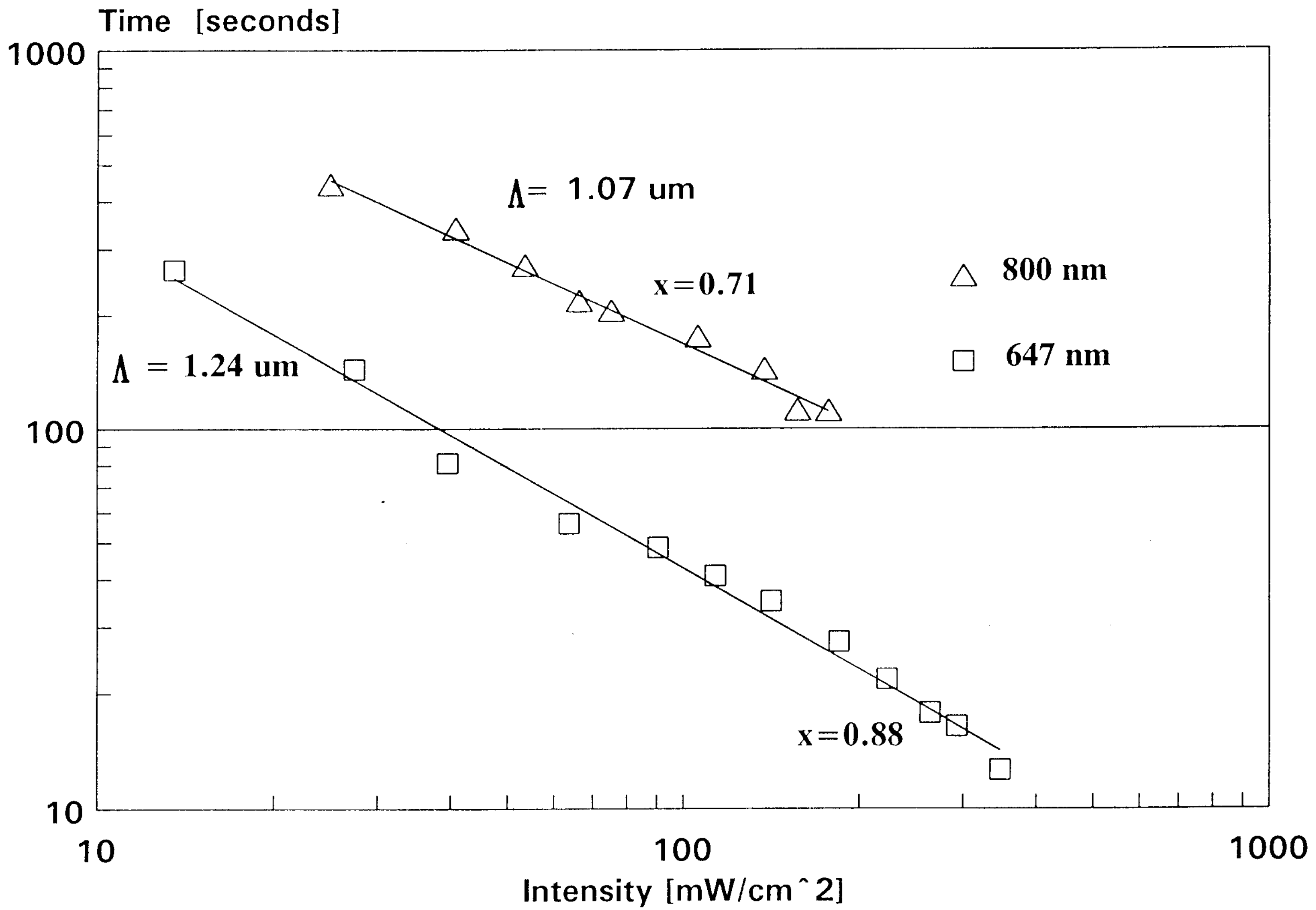
Figure captions

Figure 1	Two-beam coupling build-up time in Rh:BaTiO$_3$ at 647 nm and 800 nm. Squares: 647 nm, Triangles: 800 nm.
Figure 2	The erasure rates in undoped and Rh:BaTiO$_3$. Gratings ($\Lambda = 1.24\mu m$) written with 647 nm and probed by a He–Ne beam, and a white light source used for erasing gratings.
Figure 3	Schematic diagram of double phase conjugation experiment. I_1 and I_2: input beams, I_3: one of the phase conjugate beams.
Figure 4	Double phase conjugation at 736 nm as a function of the input beams intensity ratio.
Figure 5	Double phase conjugation at 760 nm as a function of the input beams intensity ratio.
Figure 6	Double phase conjugation at 800 nm as a function of the input beams intensity ratio.
Figure 7	Maximum phase conjugate transmission as a function of input beams' wavelength.
Figure 8	Double colour pumped oscillation between 647 nm and 800 nm as a function of the input beams intensity ratio.


Time [seconds]
1000
100
10
10
100
1000
Intensity [mW/cm^2]
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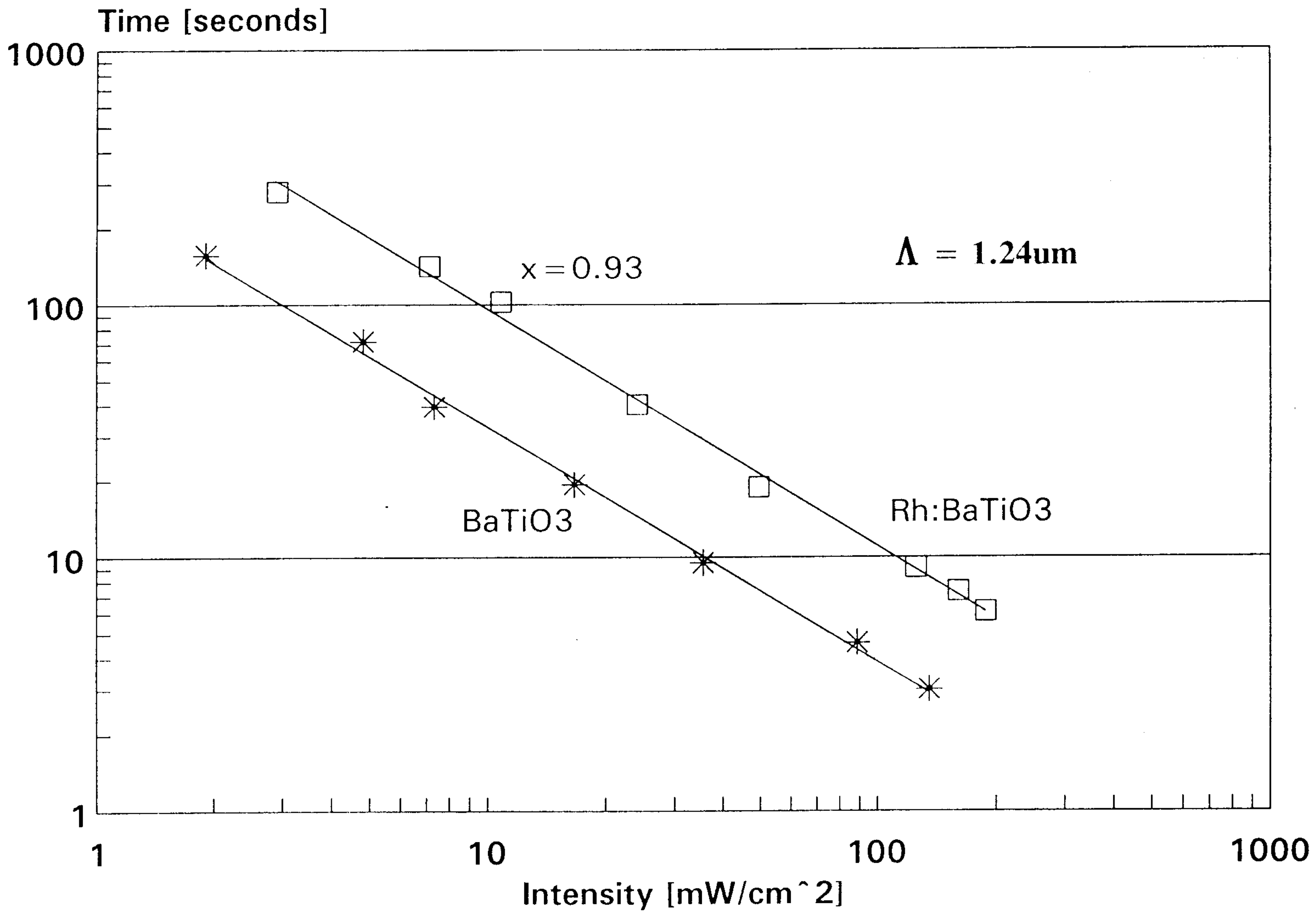
x=0.71
Λ = 1.24 um
x=0.88
△ 800 nm
□ 647 nm


Time [seconds]
1000
100
10
1
1
10
100
1000
Intensity [mW/cm^2]
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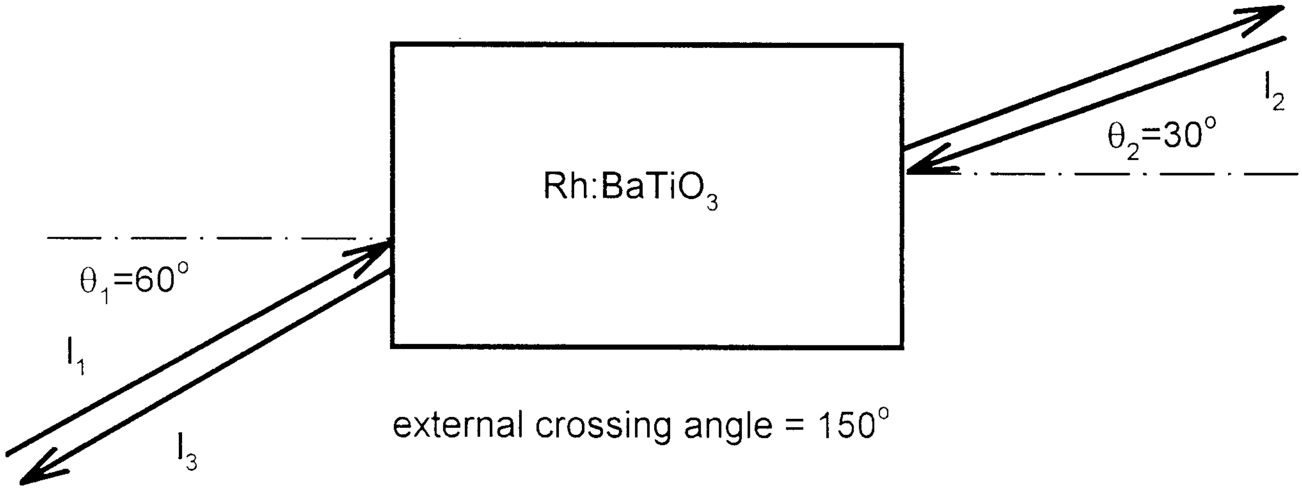
Λ = 1.24um
BaTiO3
Rh:BaTiO3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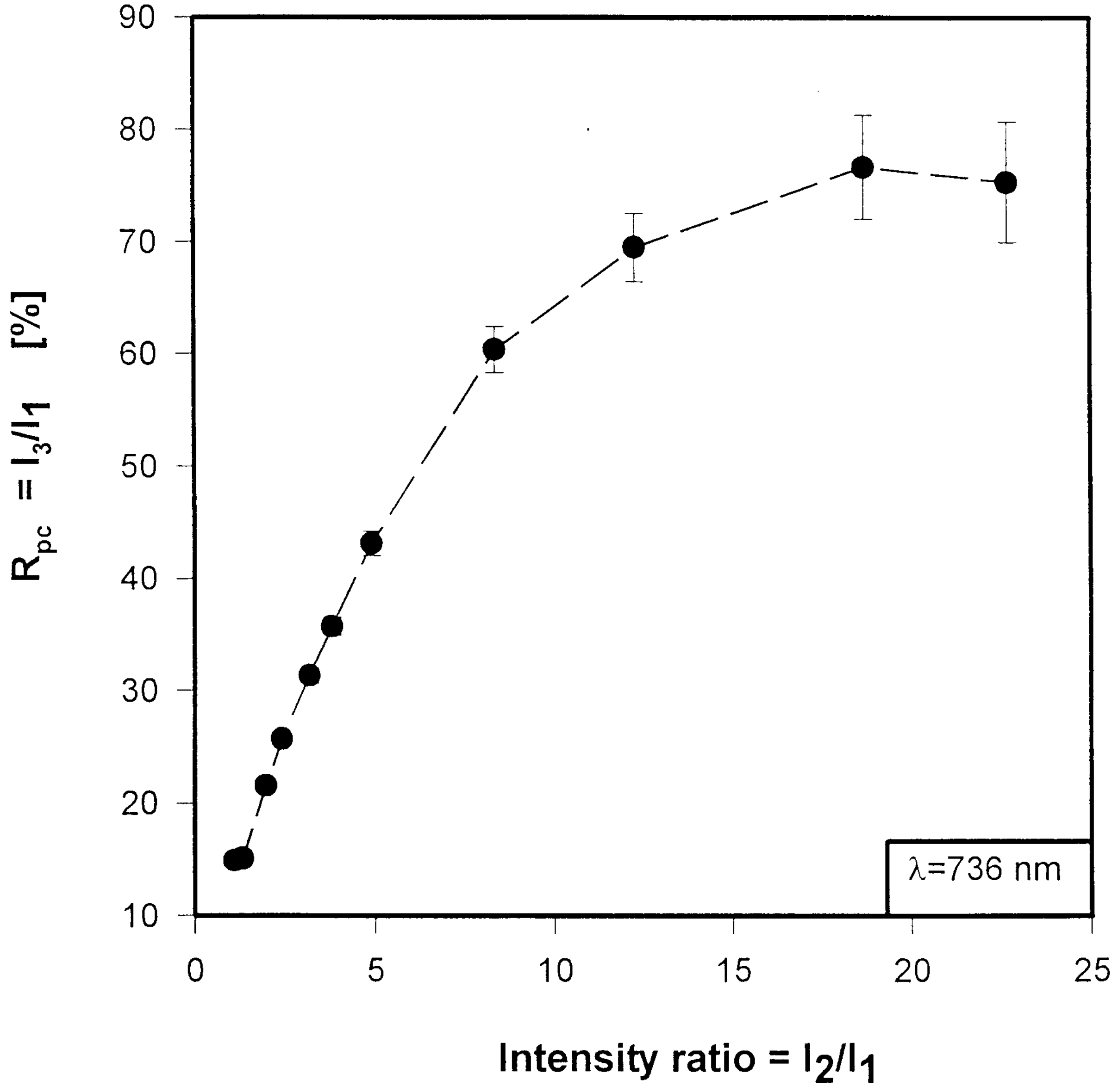
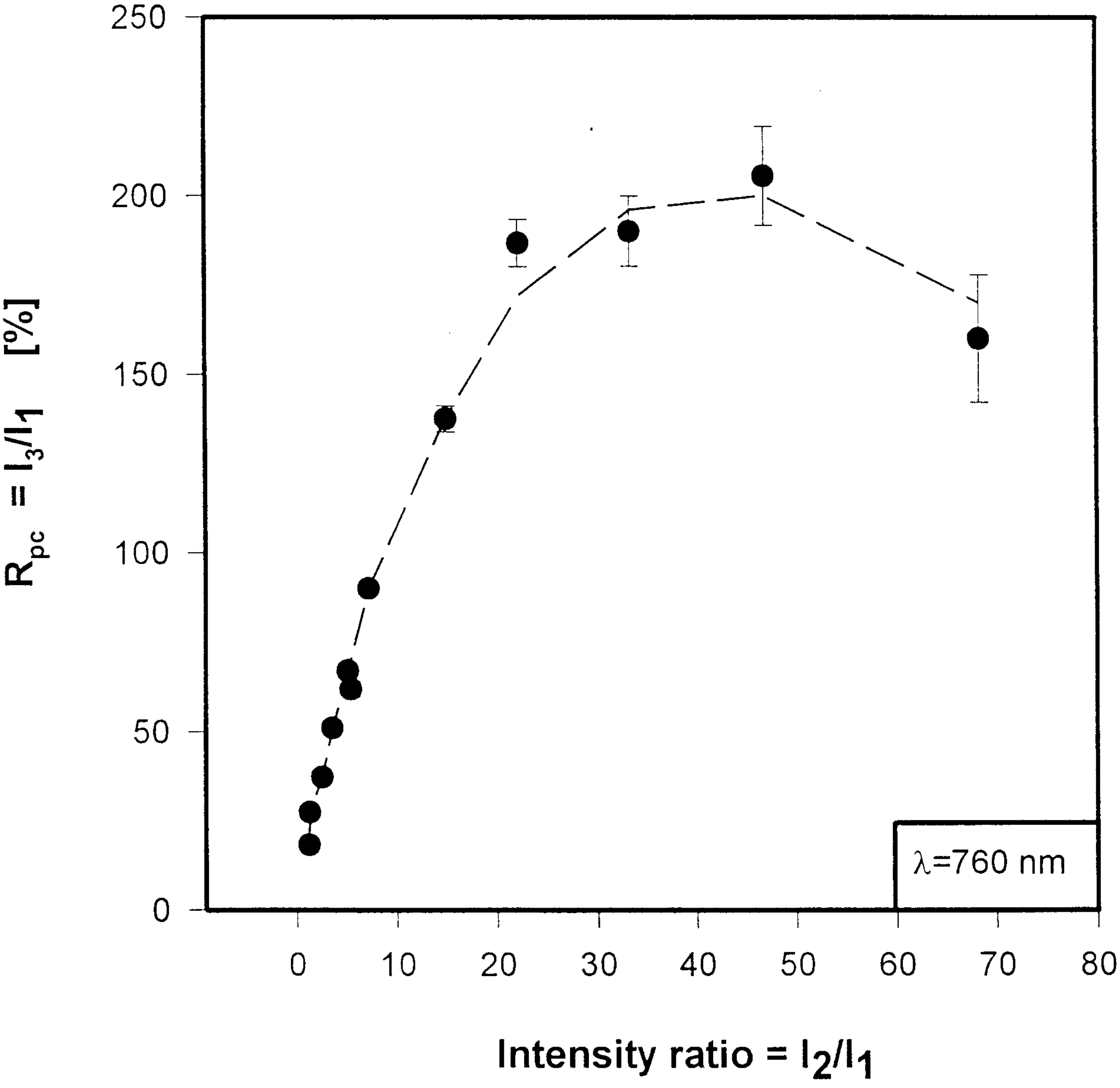
θ_1=60°
θ_2=30°
I_1
I_2
I_3
external crossing angle = 150°

Fig. 3



90
80
70
60
50
40
30
20
10
$R_{pc} = I_3/I_1$ [%]
λ=736 nm
0
5
10
15
20
25
Intensity ratio = I_2/I_1

Fig. 4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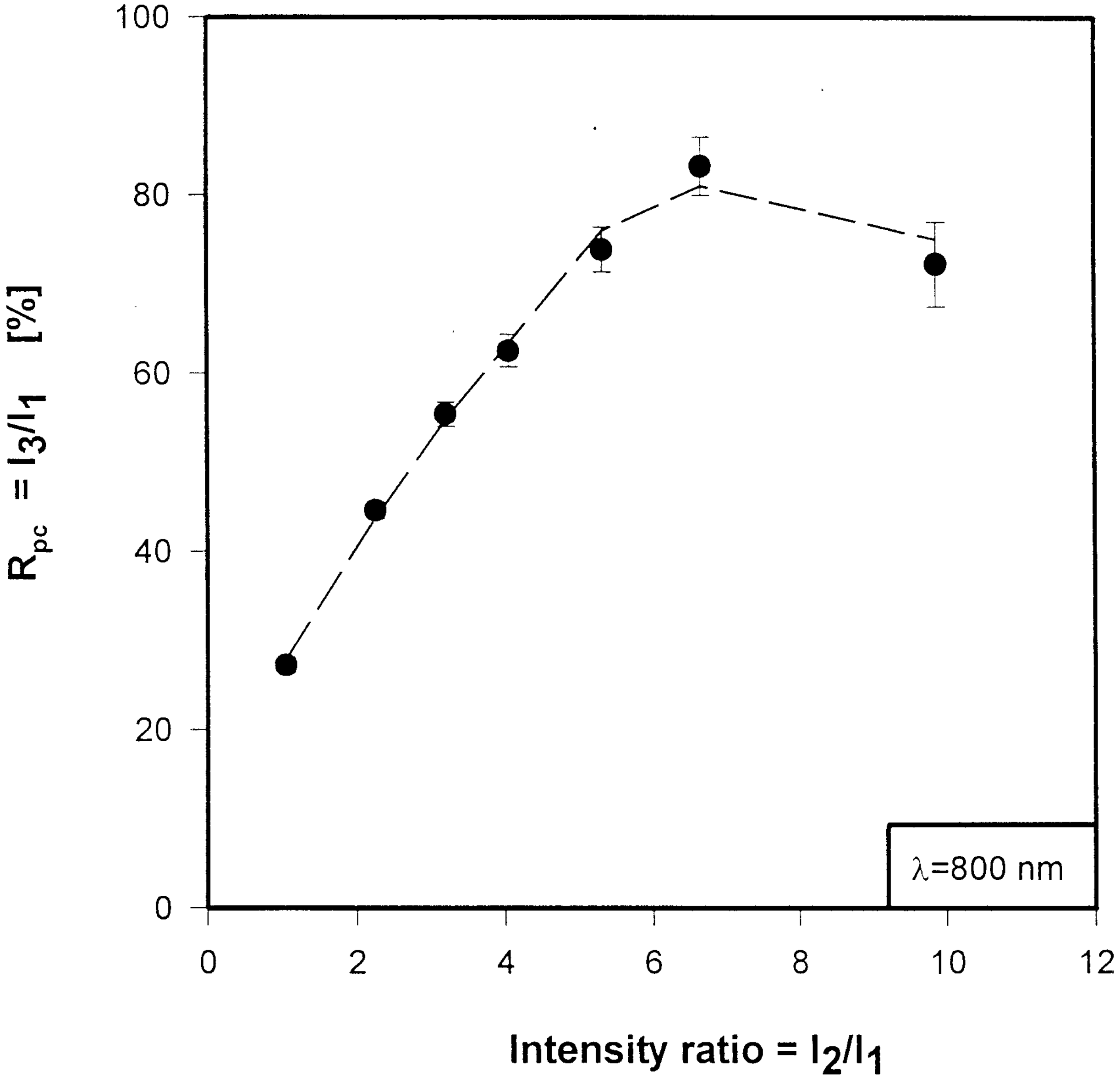
200
150
100
50
0
0
10
20
30
40
50
60
70
80
$R_{pc} = I_3/I_1$ [%]
Intensity ratio = I_2/I_1
λ=760 nm

Fig. 5


100
80
60
40
20
0
$R_{pc} = I_3/I_1$ [%]
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2
4
6
8
10
12
Intensity ratio = I_2/I_1
λ=800 nm

Fig. 6

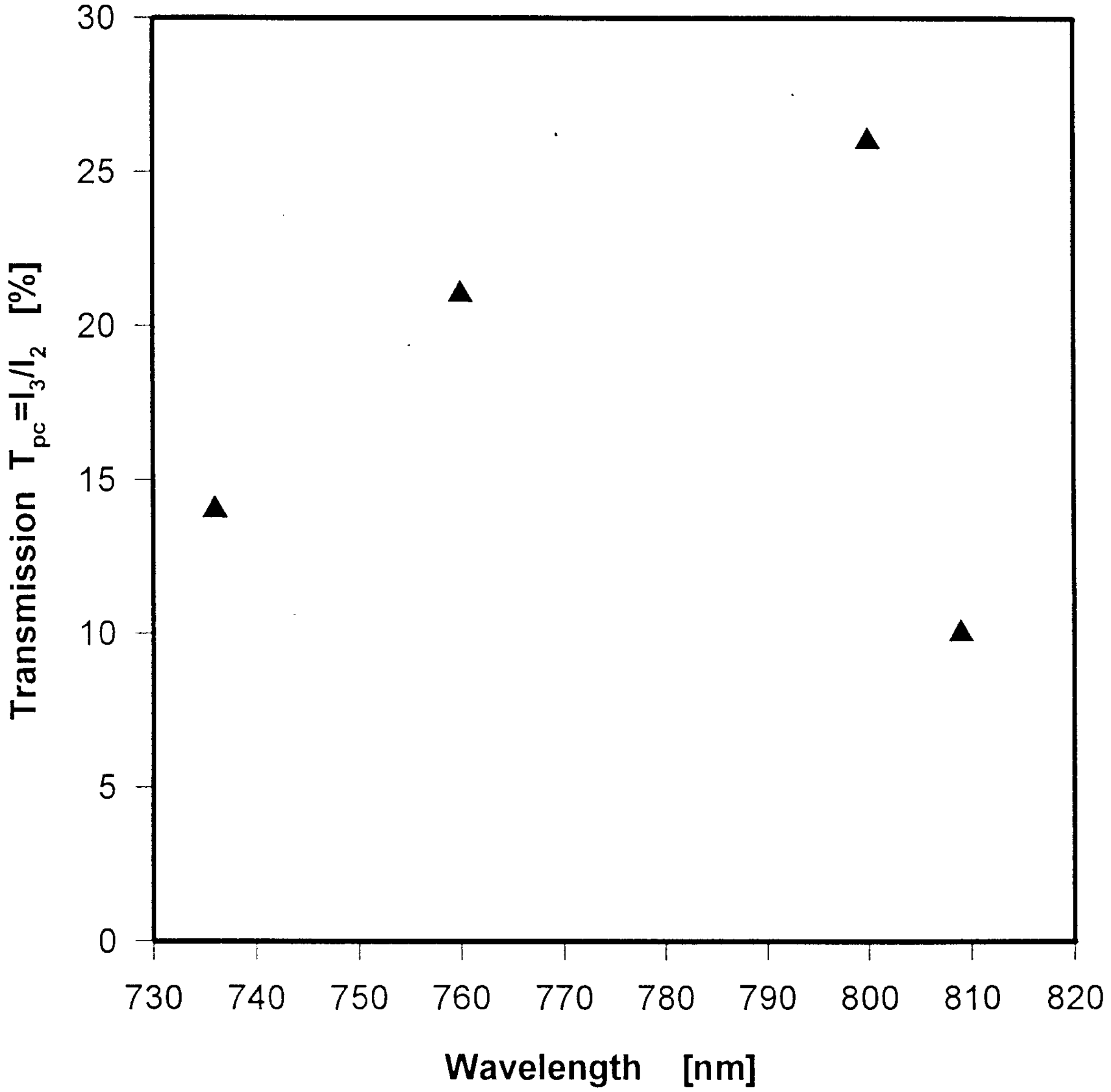
30
25
20
15
10
5
0
Transmission $T_{pc}=I_3/I_2$ [%]
730
740
750
760
770
780
790
800
810
820
Wavelength [nm]

Fig. 7

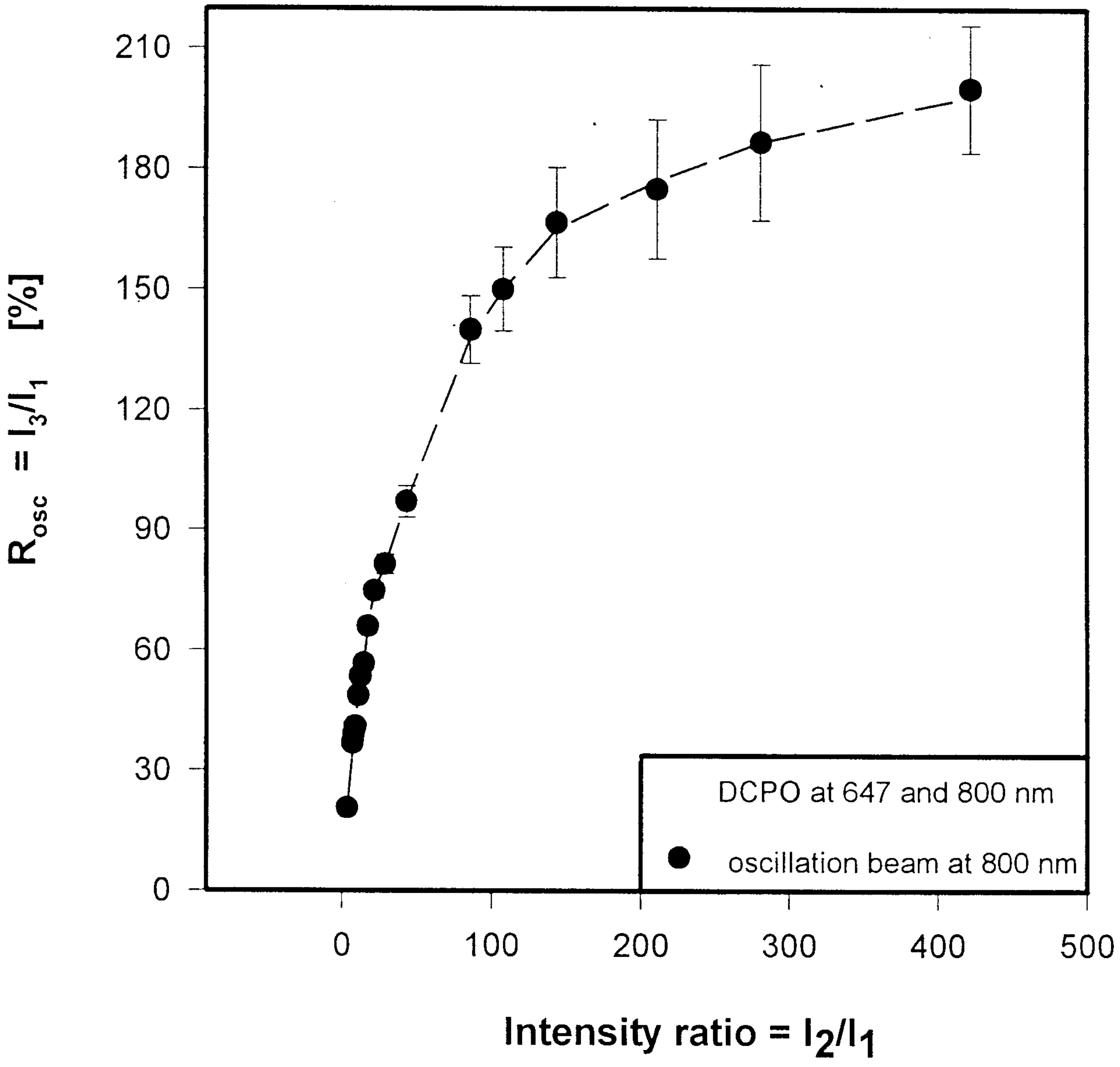
210
180
150
120
90
60
30
0
$R_{osc} = I_3/I_1$ [%]
0
100
200
300
400
500
Intensity ratio = I_2/I_1
DCPO at 647 and 800 nm
● oscillation beam at 800 nm

Fig. 8