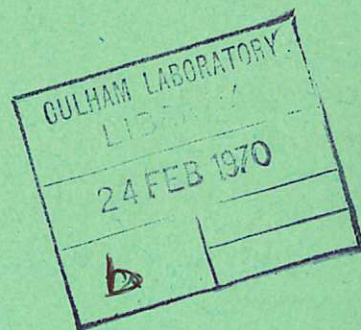


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BRAGG REFLECTION FROM A PHASE GRATING INDUCED BY NON-LINEAR OPTICAL EFFECTS IN LIQUIDS

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1969

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by

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A B S T R A C T

Bragg reflection of light from a phase grating has been used as a technique for the investigation of non-linear optical phenomena. A continuous argon ion laser was used to probe the structure induced in a liquid by the non-linear interaction of a high intensity ruby laser beam with that liquid. This arrangement had the advantage of allowing direct space and time resolved measurements of the induced effects.

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1. INTRODUCTION

The strong electric field of the light output of a ruby laser affects the refractive index of a medium through which it passes in a number of ways. The most significant of these are:-

1. Electrostriction
2. Kerr effect
3. Thermal effects - in absorbing media.

When the light is reflected back along its own path, large oscillating fields exist at the antinodes of the resulting standing wave. As the above effects are dependent on the square of the field a spatially periodic variation of refractive index is set up proportional to the mean square of the local field (Fig.1). If the frequency of the back-reflected light is slightly shifted, the nodes and antinodes will propagate through the medium. The distortion associated with each of the above effects also has a characteristic velocity of propagation. (Electrostriction gives acoustic phonons - the Kerr effect gives optical phonons - the thermal fluctuations decay but do not propagate.) When the velocity of propagation of the nodes of the field matches that of the distortion of the medium, the effect on the medium has a resonant maximum. (It is this situation which, at sufficiently high laser powers, can give rise to the stimulated scattering associated with each of the interactions, i.e. electrostriction gives stimulated Brillouin scattering^(1,2), the Kerr effect gives stimulated Rayleigh Wing^(3,4) and stimulated Raman scattering⁽⁵⁾, the thermal effect gives stimulated thermal Rayleigh scattering^(6,7)).

Thus by choosing the appropriate feedback frequency we can select the form of interaction we wish to investigate.

The refractive index variation set up in this way, by the ruby laser light, acts as a phase grating^(8,9,10) upon a probing argon laser beam traversing the medium. Each layer of high and low index reflects a small portion of the incident argon light. When these portions add in phase, a maximum overall reflectivity is reached. The condition for this is the Bragg condition:

$$\theta_0 = \cos^{-1} \frac{\lambda_a}{\lambda_r} \cdot \frac{n_r}{n_a} = 45.5^\circ$$

where θ_0 is the angle between the two beams, and suffices a and r refer to the argon and ruby wavelengths respectively. At this angle, providing the reflectivity is small

$$\text{reflectivity} = \left(\frac{\pi N \delta}{2 \cos^2 \theta} \right)^2 \quad (\text{see Appendix})$$

where N = number of modulations crossed

$$\delta = \frac{\Delta n_a}{2 n_{ao}}$$

Δn_a = difference between maximum and minimum refractive index (for argon light) induced by the standing wave.

n_{ao} = refractive index of the undisturbed medium for argon light.

When $\Delta n \sim 2 \times 10^{-6}$ and length of interaction region ~ 1 cm

reflectivity $\sim 1\%$.

The angular width at half reflectivity is

$$\frac{4 \times 1.39}{\pi N \tan \theta_0} \quad (\text{see Appendix})$$

For 1 cm interaction length

angular width $\sim 5 \times 10^{-5}$ radians ~ 10 seconds.

2. EXPERIMENTAL

Fig.2 shows the experimental arrangement. Single longitudinal mode output of the ruby laser was achieved by the use of two narrow dye cells for Q-switching and a resonant reflector as the output mirror. An aperture was used to cut down the divergence of the beam, and power densities of up to 100 MW/cm^2 were transmitted. The pulse duration was of the order of 15 nsec.

The argon laser gave 1 W output at 4880 \AA with a beam divergence of 1 mrad. Accurate adjustment of the Bragg angle between the argon light and ruby induced structure was achieved by sensitive control of mirror M_1 . A Fabry-Perot interferometer was used to check the single mode output of the ruby laser and to measure the frequency shift of the backward-going beam. These beams were distinguished by the use of the $\lambda/4$ plate and polaroid quadrant⁽⁷⁾, as shown in the diagram. The reflected beam could either be shifted or unshifted in frequency. An unshifted frequency was obtained simply by the use of a mirror normal to the ruby beam. A shifted frequency was generated by stimulated back-scattering of light focussed into cell (2), as shown in the diagram. The frequency shift was determined by the liquid contained in cell (2).

An advantage of this set up was that the detectors D_1 , D_2 measured the incident ruby beam in each direction while P and D_3 simultaneously measured the amplitude of the refractive index modulation in the cell and the amplification which it caused in the backward beam. This was achieved by the use of cable delay lines allowing simultaneous display of four pulses on a single trace of a Tectronix 454 oscilloscope.

The intensities of the beams were controlled (and feed-back into the laser minimised) by the use of CuSO_4 attenuators. The Bragg angle of 45.5° between the ruby and argon laser beams permits the very simple geometry of cell (1) and normal incidence of the argon beam as shown. Spatial resolution could be directly achieved by longitudinal movement of the cell along the ruby beam.

3. RESULTS

The experimental investigations were carried out mainly in absorbing media using unshifted frequency feedback. The thermal effect was therefore dominant.

(a) Angular Dependence

Using a solution of copper acetate in methanol with absorption coefficient $\alpha = 0.15 \text{ cm}^{-1}$ the angle giving maximum Bragg reflection was measured to be $45.5 \pm 0.1^\circ$, and was equal to the theoretical value given by $\cos^{-1} \frac{\lambda_a}{\lambda_r} \cdot \frac{n_r}{n_a}$. While the angle could only be measured absolutely to $\pm 0.1^\circ$, small incremental adjustments could be made much more accurately⁽¹¹⁾. The angular width at half reflectivity was found to be ~ 5 minutes corresponding to the divergence of the argon laser beam.

(b) Magnitude of Reflectivity

The refractive index modulation due to thermal effects is given by⁽¹²⁾

$$\Delta n = \gamma \frac{P}{2\ell\chi(n_r k_r)^2} \left(\frac{\partial n}{\partial T} \right)$$

where γ is a numerical factor such that $0.5 < \gamma < 1$ and P is the power per unit area absorbed in the medium of length ℓ and thermal conductivity χ .

Hence with the same solution as in (a), with an intensity of 23 MW/cm² and $\ell \sim 4$ cm

$$\Delta n \sim 4 \times 10^{-6}$$

Now reflectivity = $\left(\frac{\pi N \delta}{2 \cos^2 \theta} \right)^2 \sim 4\%$ for argon light incident at the Bragg angle. However, the theoretical angular width at half reflectivity of the Bragg is of the order of 10 seconds for a non-divergent beam (see Introduction), whereas the divergence of the argon laser beam was about 5'. Hence only about 3% of the argon beam was available for reflection, and thus resultant reflectivity $\sim 0.1\%$.

This estimate was experimentally confirmed.

(c) Dependence of Reflectivity on Ruby Laser Power

The change in refractive index at any point is proportional to the mean square of the local field. Let \underline{E}_1 and \underline{E}_2 be the electric vectors of the forward- and backward- going beams respectively.

Now

$$E_1 = A_1 \sin(kz - \omega t)$$

$$E_2 = A_2 \sin(-kz - \omega t).$$

The local field $E = E_1 + E_2$

$$= A_1 \sin(kz - \omega t) + A_2 \sin(-kz - \omega t)$$

$$\bar{E}^2 = \frac{1}{2}A_1^2 + \frac{1}{2}A_2^2 + \frac{A_1 A_2}{2} \cos 2kz$$

therefore the difference between maxima and minima of the mean square of the local field = $A_1 A_2$.

Hence the difference between the maxima and minima of the refractive index $\delta n \propto A_1 A_2$.

Now reflectivity of grating $\propto (\delta n)^2$ (see Introduction)

$$\propto I_1 I_2 \quad \text{where } I_1, I_2 \text{ are the intensities of the forward- and backward- going beams respectively. Fig.3}$$

shows the dependence of reflectivity of a solution of copper acetate in methanol ($\alpha = 0.15 \text{ cm}^{-1}$) on I_1 when $I_2 = 0.33 I_1$. In this case reflectivity $\propto I_1^2$ as is confirmed by the graph. We ascribe the errors mainly to variation of the divergence of the ruby light due to lack of transverse mode control.

Fig.4 shows that the reflectivity is proportional to I_2 when I_1 is kept constant. These two results confirm the theoretical relationship.

Further power dependence investigations were made in the case of a saturable absorber (cryptocyanine in methanol). As was expected, the I_1^2 dependence breaks down at about 10 MW/cm^2 owing to the bleaching of the dye^(13,14,15) (Fig.5).

(d) Dependence of Reflected Power on Incident Argon Laser Power

To ensure that the argon light was acting solely as a probe and not contributing significantly to the non-linear effects, the dependence of reflected power on the incident argon power was investigated. For solutions not absorbing at the argon wavelength, e.g. copper acetate in methanol, the expected linear dependence was observed (Fig.6). However, for solutions having even a slight absorption at the argon wavelength, e.g. nitrobenzene, thermal de-focusing⁽¹⁶⁾ of the argon beam greatly reduced the Bragg reflection.

(e) Dependence of the Reflectivity on Concentration

The reflectivity of the grating was also investigated as a function of the absorption coefficient. As expected, different solutes in the same solvent gave equal reflectivities when the absorption coefficients of the solutions were equal.

The measurement of the reflection as a function of the absorption coefficient α was complicated by the fact that variation of α affects the values of E_1 and E_2 . This can only be corrected for as long as the non-linear effect in the cell has a negligible effect on the ruby beam, a condition that requires a much shorter cell than that used in this experiment.

(f) Time Resolution

Each of the non-linear effects mentioned earlier has a characteristic relaxation time⁽¹⁷⁾. These have the following orders of magnitude:

Electrostriction	- 10^{-9} to 10^{-10} sec
Kerr effect	- 10^{-11} to 10^{-12} sec
Thermal effect	- 10^{-7} to 10^{-8} sec.

The length of the ruby pulse (~ 15 nsec) and the resolution of the instruments (~ 7 nsec) only allowed investigation of the thermal relaxation.

Fig.7 shows the time profile of the ruby laser pulse and the resulting Bragg reflected pulse in the thermal and the electrostrictive cases. The relaxation time of the effect of electrostriction is very fast (compared with the pulse profile and instrument resolution). Hence the profile of this pulse provides a measure of the resolution of the instruments, (7 nsec). The relaxation time of a thermal grating is calculated from the bulk properties of the medium by the formula⁽⁷⁾

$$\tau = \frac{c\rho}{\chi(2k_r n_r)^2}$$

where c = specific heat at constant pressure and ρ = density. This gives a value of 16.5 nsec for methanol. The points marked on Fig.7

are the theoretical points given this relaxation time and the resolution time of the instruments. There is good agreement for this value of relaxation time and significant misfit if it is varied by more than 2 nsec.

Similar agreement was obtained in the case of a number of other liquids.

This technique can be used more generally for investigation of any non-linear optical effect dependent on E^2 . In particular short pulses and better detector resolution will allow direct measurement of the lifetimes of the phonons generated by the stimulated Brillouin effect.

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APPENDIX

Let θ = angle of incidence at a refractive index boundary

θ' = angle of refraction at a refractive index boundary

A_i = amplitude of wave incident on boundary

δA_r = amplitude of wave reflected from boundary

δn = very small change of refractive index at boundary

then $\frac{\delta A_r}{A_i} = \frac{-\sin(\theta-\theta')}{\sin(\theta+\theta')} \sim \frac{-\delta n}{2n_{ao}\cos^2\theta}$ for parallel polarisation

$$\text{in limit } \frac{1}{\delta n \rightarrow 0} \cdot \frac{dA_r}{dn} = \frac{-1}{2n_{ao}\cos^2\theta}$$

Now let

n_{ao} = undisturbed refractive index for argon light

n_{ro} = undisturbed refractive index for ruby light

Δn_a = difference between max and min refractive index for argon light

λ_a = wavelength of argon light in vacuo

λ_r = wavelength of ruby light in vacuo

$$k_a = \frac{2\pi}{\lambda_a}$$

$$k_r = \frac{2\pi}{\lambda_r}$$

z = distance along ruby beam direction

$$\text{then } n_a = n_{ao} + \frac{\Delta n_a}{2} \sin 2k_r n_{ro} z$$

$$\frac{dn_a}{dz} = \frac{\Delta n_a}{2} 2k_r n_{ro} \cos(2k_r n_{ro} z)$$

$$\therefore \frac{1}{A_i} \cdot \frac{dA_r}{dz} = \frac{-\Delta n_a \cdot k_r n_{ro} \cos(2k_r n_{ro} z)}{2n_{ao} \cdot \cos^2\theta}.$$

Now light reflected from boundary at z has phase lag of

$$2n_{ao} k_a z \cos\theta \quad \text{at } z = 0$$

$$\therefore \frac{1}{A_i} \frac{dA_{ro}}{dz} = \frac{-\Delta n_a \cdot k_{rro}}{2n_{ao} \cdot \cos^2\theta} \cdot \cos(2k_{rro} z) \cdot e^{i2n_{ao} k_a z \cos\theta}$$

where A_{ro} is the amplitude at $z = 0$ of the wave reflected from a depth z of grating. Then total amplitude, A_R , of wave at $z = 0$ reflected from grating of depth L is given by

$$\frac{A_R}{A_i} = \int_0^L \frac{1}{A_i} \cdot \frac{dA_{ro}}{dz} \cdot dz = \frac{-\Delta n_a k_{rro}}{2\cos^2\theta n_{ao}} \int_0^L \cos(2k_{rro} z) e^{i2n_{ao} k_a z \cos\theta} dz$$

assuming $A_i \sim \text{const} = A_r$ i.e. total reflectivity $\ll 1$ where

L = total length of modulations probed.

Now let $2k_{rro} = k$

$$2k_a n_{ao} \cos\theta = k'$$

$$\left(\frac{A_R}{A_i} \right) = \frac{-k \Delta n_a}{4 \cos^2\theta n_{ao}} \int_0^L \cos kz e^{ik' z} dz$$

$$\begin{aligned} \left(\frac{A_R}{A_i} \right) &= \frac{-k \Delta n_a}{8 \cos^2\theta n_{ao}} \left[\frac{\sin(k'+k)L}{k'+k} + \frac{\sin(k'-k)L}{k'-k} + \right. \\ &\quad \left. + i \left[\frac{1-\cos(k'+k)L}{k'+k} + \frac{1-\cos(k'-k)L}{k'-k} \right] \right] \end{aligned}$$

for $k' \sim k$ terms with $k'-k$ denominator \gg terms with $k'+k$ denominator

$$\begin{aligned} \therefore \left| \frac{A_R}{A_i} \right|^2 &= \left(\frac{k \Delta n_a}{8 \cos^2\theta n_{ao}} \right)^2 \left[\frac{\sin^2(k'-k)L + (1-\cos(k'-k)L)^2}{(k'-k)^2} \right] \\ &= \left(\frac{k \Delta n_a L}{8 \cos^2\theta n_{ao}} \right)^2 \left[\frac{\sin\left(\frac{k'-k}{2}\right) L}{\left(\frac{k'-k}{2}\right) L} \right]^2 \end{aligned}$$

This has maximum when $k' = k$

i.e. when $\cos \theta = \frac{\lambda_a}{\lambda_r} \cdot \frac{n_{ro}}{n_{ao}}$

then $\frac{I_R}{I_I} = \left| \frac{A_R}{A_I} \right|^2 = \left(\frac{k \Delta n_a L}{8 \cos^2 \theta n_{ao}} \right)^2$

where I_R, I_I are the reflected and incident intensities

$$\text{reflectivity} = \frac{I_R}{I_I} = \left(\frac{\pi N \delta}{2 \cos^2 \theta} \right)^2$$

where $N = \frac{2n_{ro} L}{\lambda_r}$

$$\delta = \frac{\Delta n_a}{2n_{ao}}$$

At half reflectivity $\frac{\sin\left(\frac{k' - k}{2}\right) L}{\left(\frac{k' - k}{2}\right) L} = \frac{1}{\sqrt{2}}$

now $k' - k = 2k_a n_{ao} \cos \theta - 2k_r n_{ro}$

let $\theta = \theta_0 + \delta\theta$ where $\theta_0 = \cos^{-1} \frac{k_r n_{ro}}{k_a n_{ao}}$

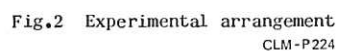
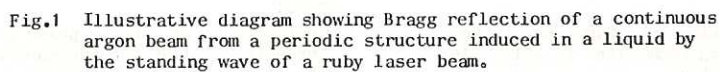
$$k' - k = -\delta\theta \sqrt{k_a^2 n_{ao}^2 - k_r^2 n_{ro}^2}$$

$$\therefore \frac{\sin\left(\frac{\delta\theta}{2} \sqrt{k_a^2 n_{ao}^2 - k_r^2 n_{ro}^2} \cdot L\right)}{\frac{\delta\theta}{2} \sqrt{k_a^2 n_{ao}^2 - k_r^2 n_{ro}^2} \cdot L} = \frac{1}{\sqrt{2}}$$

$$\therefore \frac{\delta\theta}{2} \sqrt{k_a^2 n_{ao}^2 - k_r^2 n_{ro}^2} \cdot L = \pm 1.39 \text{ radians}$$

$$\therefore \delta\theta = \pm \frac{2 \times 1.39}{\pi N \sqrt{\left(\frac{k_a n_{ao}}{k_r n_{ro}}\right)^2 - 1}}$$

$$\therefore \text{angular width at half reflectivity} = \frac{4 \times 1.39}{\pi N \tan \theta_0}$$



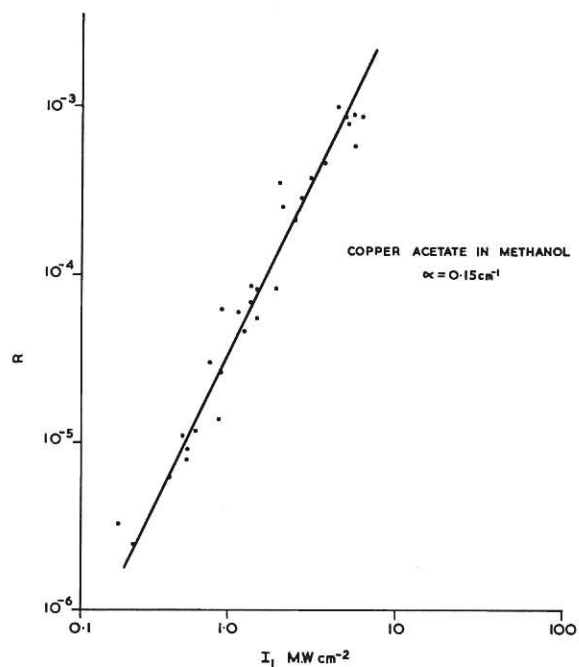


Fig.3
Reflectivity (R) of the phase grating as a function of the intensity (I_1) of the forward-going beam with $I_2 \propto I_1$. The line shows the theoretical square law.

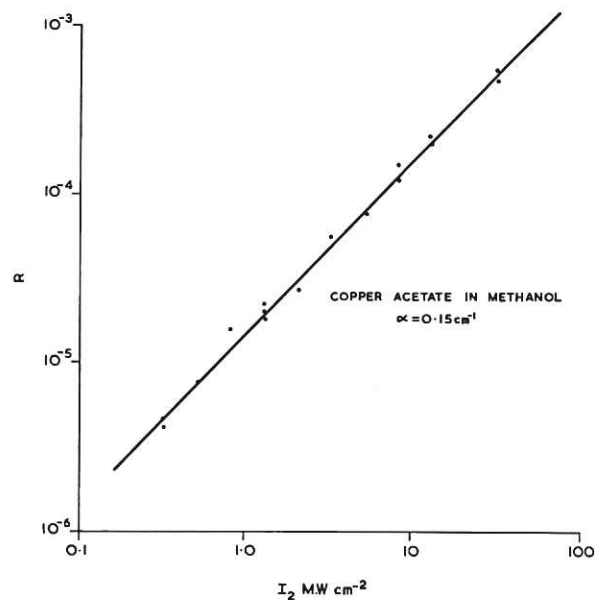


Fig.4
Reflectivity (R) of the phase grating as a function of the intensity (I_2) of the backward-going beam with I_1 constant at 71 MW cm^{-2} . The line shows the theoretical linear law.

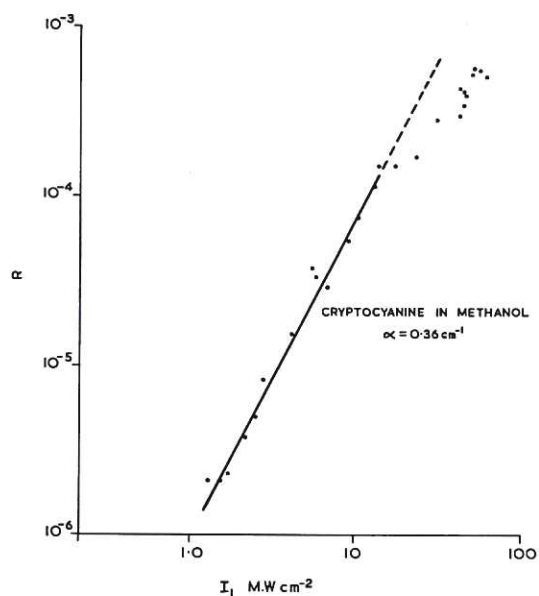


Fig.5
Reflectivity of phase grating as a function of I_1 for a saturable absorber.

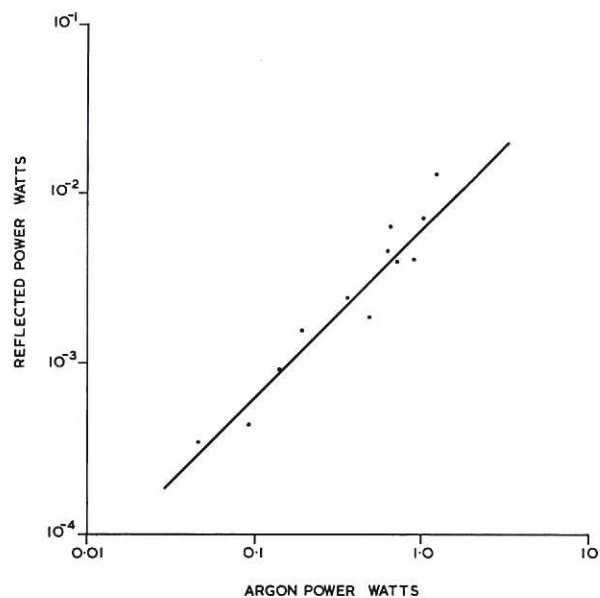


Fig.6
Reflected power as a function of incident argon laser power.

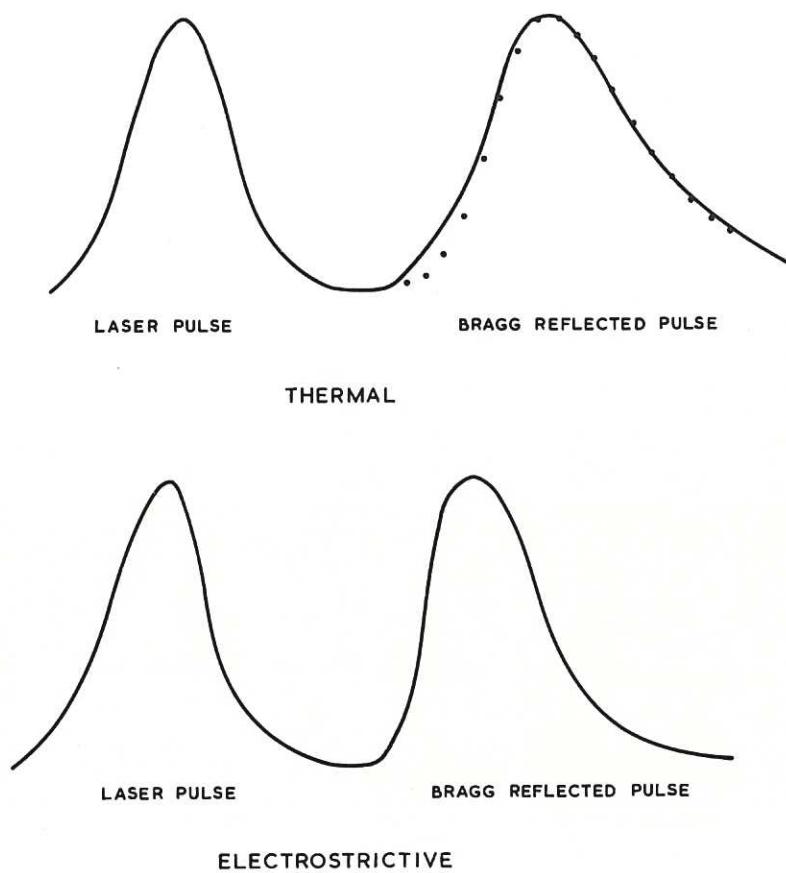
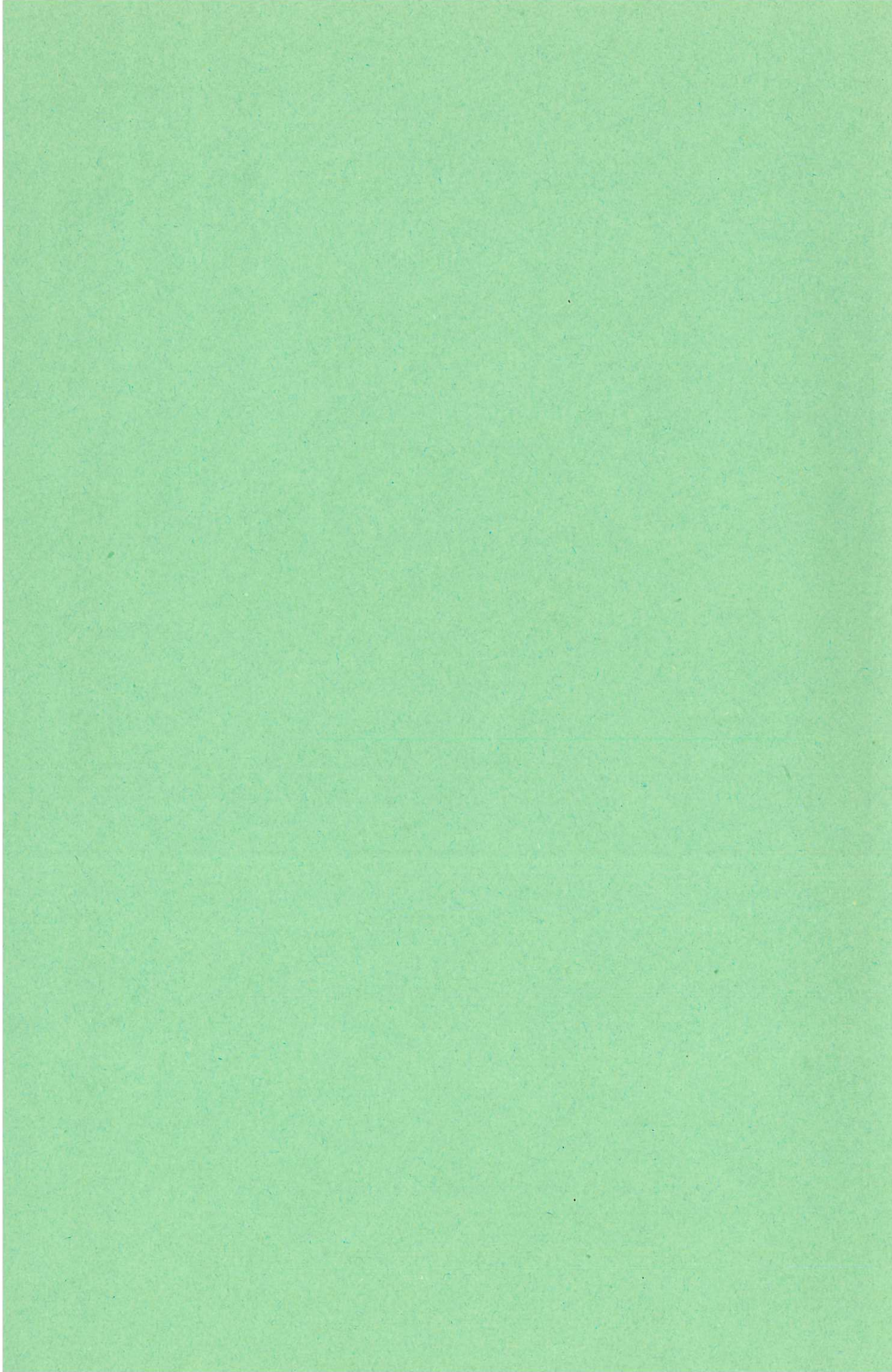


Fig.7 Time profile of the ruby laser pulse and the resulting Bragg reflected pulse in the thermal and electrostrictive cases. The points marked are those calculated given the theoretical relaxation time.

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