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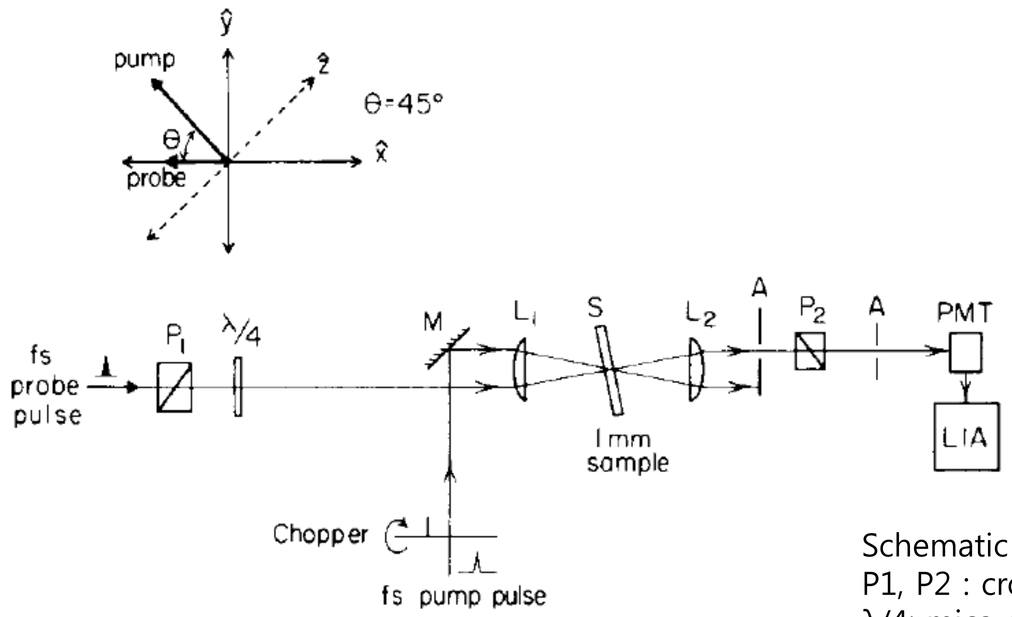
Femtosecond Optical Kerr Studies on the Origin of the Nonlinear Responses in Simple Liquids

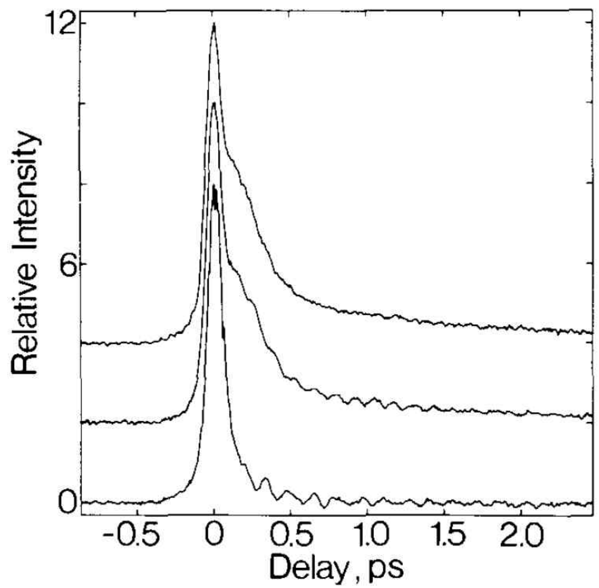
DALE McMORROW, WILLIAM T. LOTSHAW, AND GERALDINE A. KENNEY-WALLACE

Experimental

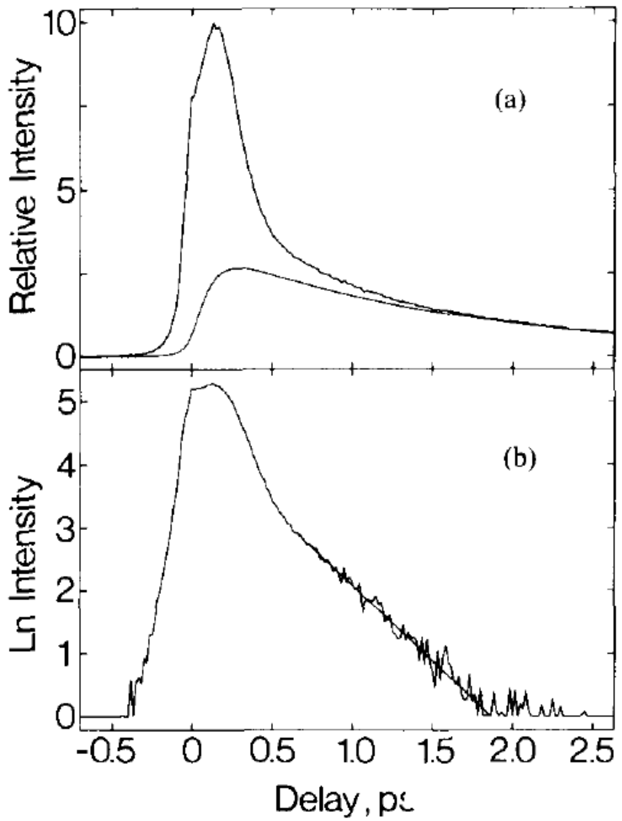


Schematic diagram of the experimental apparatus.
P1, P2 : crossed Glan-Taylor polarizer pair,
λ/4: mica quarter-wave plate,
L1 and L2: lenses,
S: sample,
A: aperture,
M: beam steering mirrors,
PMT: photomultiplier tube,
LIA: lock-in amplifier.
The relative pump and probe beam polarizations are shown in the inset with θ = 45°.

The Role of Molecular Symmetry

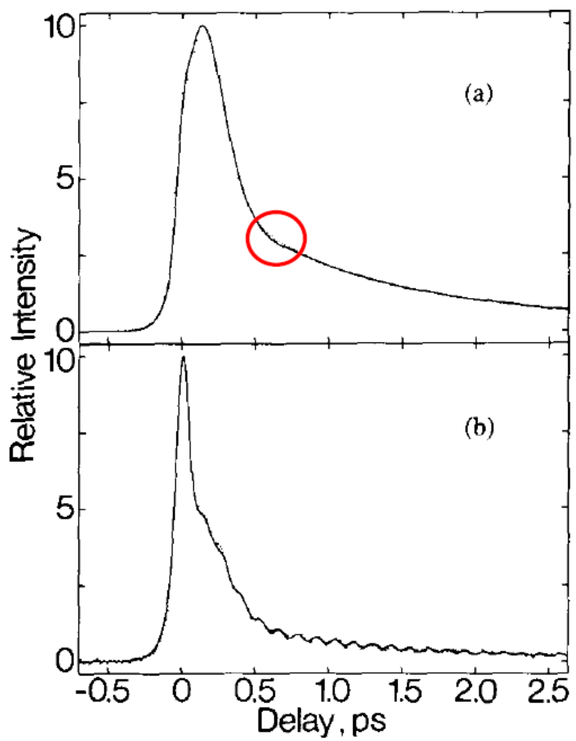


Optical Kerr signals for the liquids CH_2CI_2, $CHCl_3$, and $CC1_4$ (top to bottom).

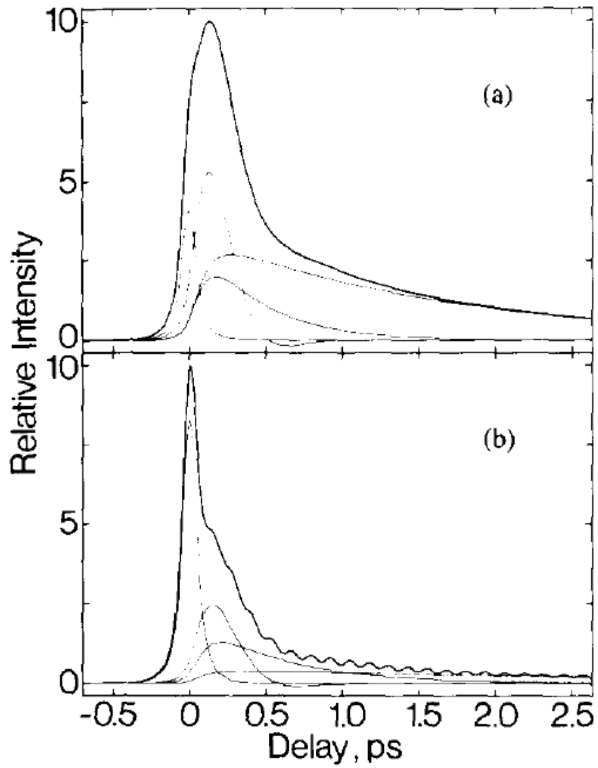


(a) Optical Kerr signal for neat CS, together with tail matched theoretical response for the diffusive orientation component, τ= 1.61 ps.

(b) Logarithmic plot of the difference of the two curves given in (a). The straight line corresponds to a time constant of 426 fs.



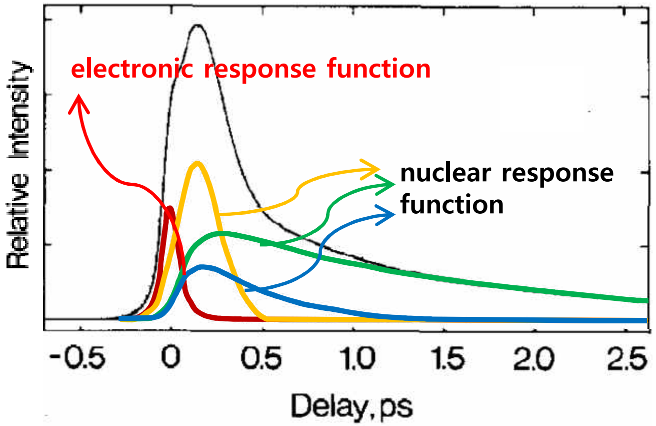
Optical Kerr signals for (a) CS_2 and (b) $CHCl_3$ (dots) shown together with theoretical best fits (solid) generated in accordance with the model described in the text.



The theoretical curves of Fig. 4 given with their respective component
curves. See text for details.

Optical kerr signal fitting

Chem. Phys. Lett. 150, 138 (1988)



- fwhm :65fs (633nm)
  sample : CS2
- $T(\tau) \propto \int_{-\infty}^{\infty} G_0^{(2)}(t) R(\tau - t) dt$

  $R(\tau) = \sigma(\tau) + r(\tau)$

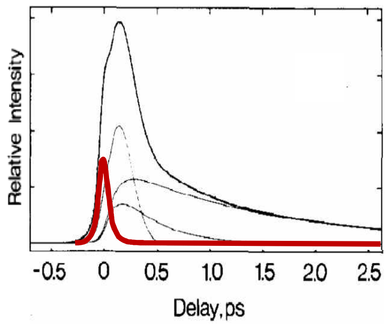
  $\sigma(\tau) = a_0 \delta(\tau)$

  $r_2(\tau) = a_2 \exp(-\frac{\tau}{\tau_{lib}}) \exp(-\frac{\alpha^2 \tau^2}{2}) \sin(\omega_0 \tau)$

  $r_3(\tau) = a_3 \exp(-\frac{\tau}{\tau_{diff}})[1 - \exp(-\frac{\tau}{\beta_3})]$

  $r_4(\tau) = a_4 \exp(-\frac{\tau}{\tau_{\text{int}}})[1 - \exp(-\frac{\tau}{\beta_4})]$

electronic response function

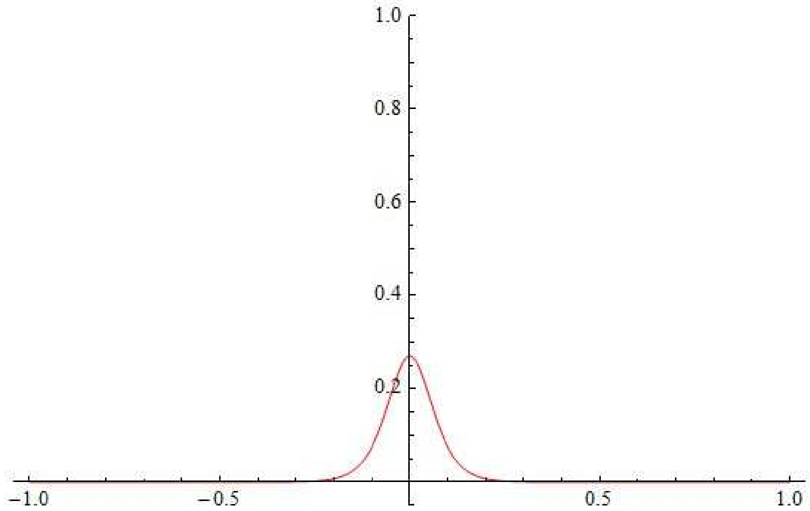



```
Clear[g2, t, τ, a0, e, s];

a0 = 0.27;
```
reference value

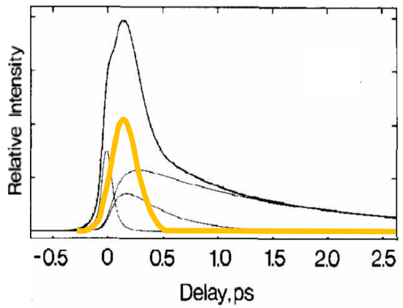
```
g2[t_] = Sech[(t)/0.0797]^2;
```
autocorrelation function

```
e[τ_] = a0 * DiracDelta[t - τ];
```
Electronic function

```
s[τ_] = ∫_{-∞}^{∞} g2[t] * e[τ] dt;

Plot[s[τ], {τ, -1, 1}, PlotRange → {-0.05, 1},
 PlotStyle → {RGBColor[1, 0, 0], PlotRange → {0, 10}}]
```



nuclear response function




```
Clear[g2, r2, sr2, a, s, t, τ];
Clear[a2, τint, α, w0];

τint = 0.329; α = 4.4; w0 = 6.67;
a2 = 0.39;
```

reference value

$$g2[t\_] = \mathrm{Sech}\left[\frac{(t)}{0.0797}\right]\text{^}2;$$

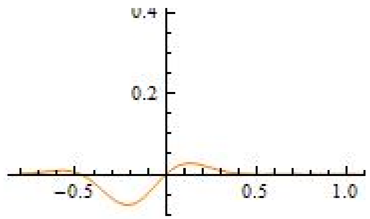
autocorrelation function

$$r2[\tau\_] = a2 * e^{-\frac{(\tau)}{\tau int}} * e^{-\frac{\alpha^2 * (\tau)^2}{2}} * \mathrm{Sin}[w0 * (\tau)];$$

nuclear response function

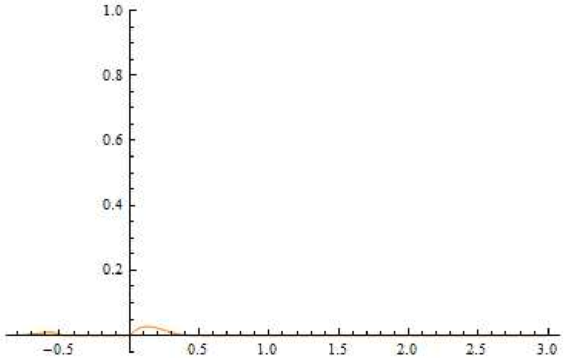
```
a[τ_] := If[r2[τ] ≥ 0, 1, 0];

sr2[τ_] = r2[τ] * a[τ];
```

$$s[\tau\_] = \int_{-\infty}^{\infty} g2[t] * sr2[\tau]\, dt$$

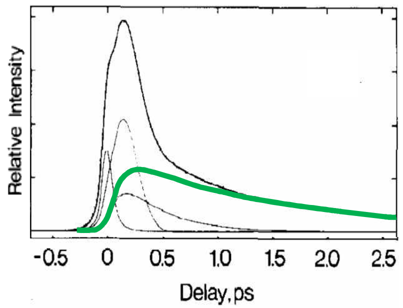



```
Plot[s[τ], {τ, -0.8, 3}, PlotStyle → {RGBColor[1, 0.5, 0]}, PlotRange → {-0.05, 1}]
```

$$0.062166\, e^{(-3.03951-9.68\,\tau)\,\tau}\, \mathrm{If}\left[1.\, e^{(-3.03951-9.68\,\tau)\,\tau}\, \mathrm{Sin}[6.67\,\tau] \geq 0, 1, 0\right] \mathrm{Sin}[6.67\,\tau]$$



nuclear response function




```
Clear[g2, r3, b, sr3, s, t, τ];
Clear[a0, a3, τdiff, cc];

a3 = 0.2; τdiff = 1.69; w0 = 6.67;
```

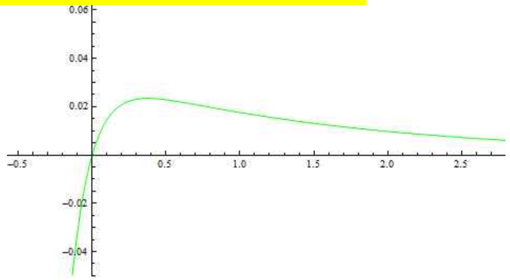
reference value

```
g2[t_] = Sech[(t)/0.05]^2;
```

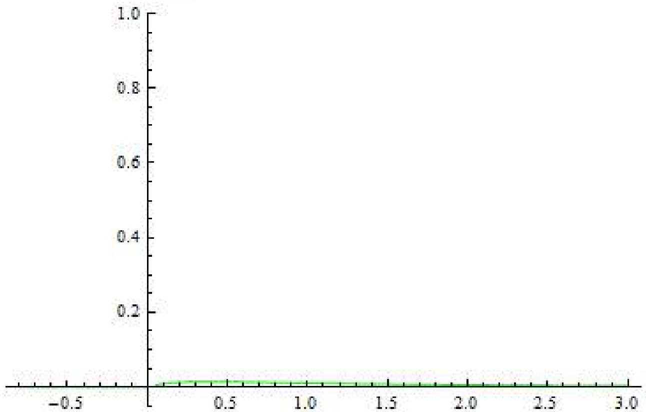
autocorrelation function

```
r3[τ_] = a3 * e^(-(τ)/τdiff) * (1 - e^(-(τ)*w0));
```

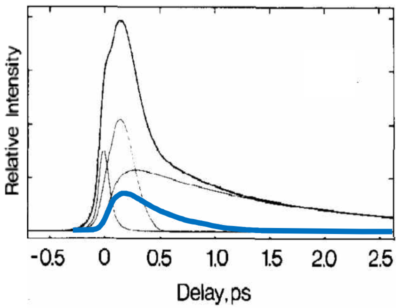
nuclear response function




```
b[τ_] := If[r3[τ] ≥ 0, 1, 0];

sr3[τ_] = r3[τ] * b[τ];

s[τ_] = ∫_{-∞}^{∞} g2[t] * sr3[τ] dt;

Plot[{s[τ]}, {τ, -0.8, 3}, PlotStyle→{RGBColor[0, 1, 0]}, PlotRange→{-0.05, 1}]
```



nuclear response function




```
Clear[g2, r4, c, sr4, s, t, τ];
Clear[a4, τint];

a4 = 0.14; τint = 0.329; w0 = 6.67;
```

reference value

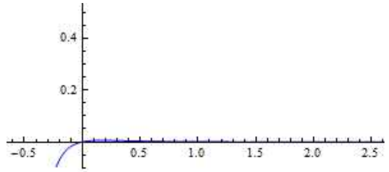
$$g2[t\_] = \mathrm{Sech}\left[\frac{(t)}{0.05}\right]^{\wedge}2;$$

autocorrelation function

$$r4[\tau\_] = a4 * e^{-\frac{(\tau)}{\tau int}} * \left(1 - e^{-(\tau) * w0}\right);$$

nuclear response function

```
c[τ_] := If[r4[τ] ≥ 0, 1, 0];

sr4[τ_] = r4[τ] * c[τ];
```



$$s[\tau\_] = \int_{-\infty}^{\infty} g2[t] * sr4[\tau]\, dt;$$

```
Plot[{s[τ]}, {τ, -0.8, 3}, PlotStyle→ {RGBColor[0, 0, 1]},
 PlotRange→ {-0.05, 1}]
```