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Femtosecond Optical Kerr Effect Studies Water

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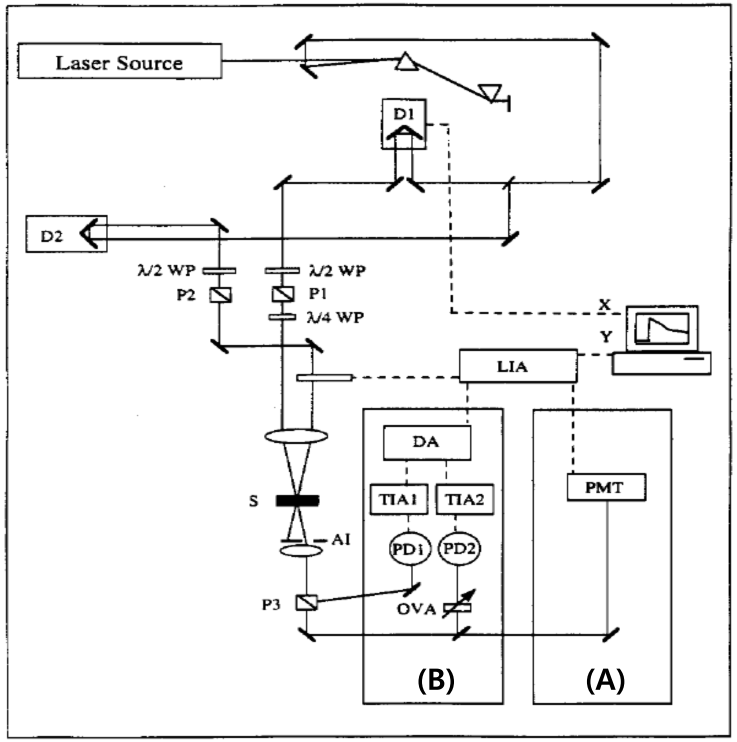
Experimental Section



- ✓ Pump : linear polarization
  Probe : 45°

  D1,D2 : optical delay

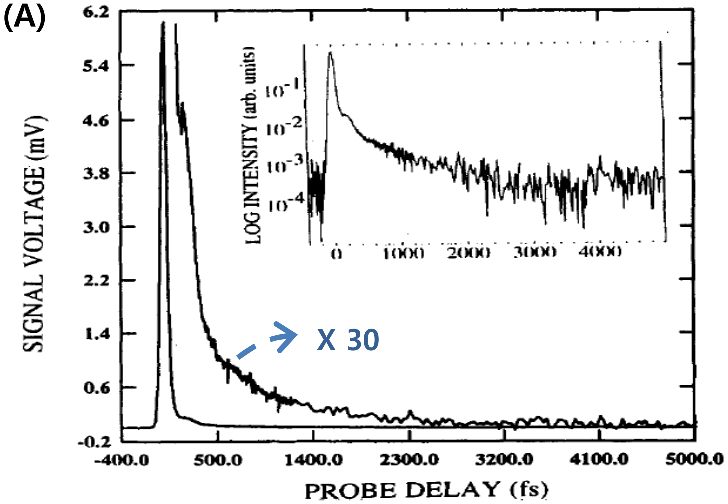
  (A) Ti:Sapphire detection sheme
  (B) amplified CPM detection sheme

  OVA : Optical variable attenuator
  PD1,PD2 : photodiode
  TIA1,TIA2 : matched transimpedance amplifiers
  DA : differential amplifier

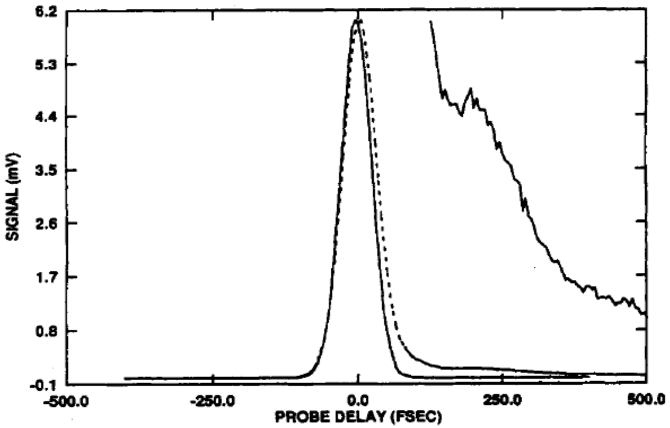
- ✓Autocorrelation : KD*P crystal (100um)

Results and Discussion

(A)



(B)

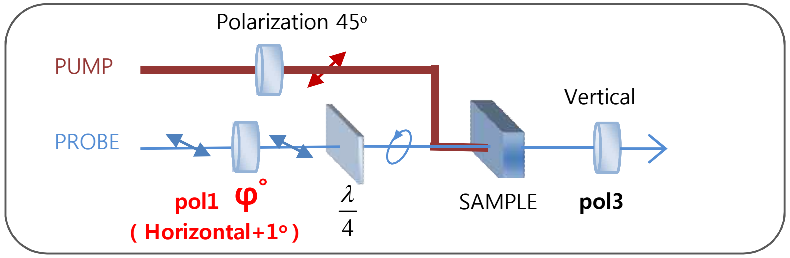


✓ **Optical Kerr effect measurement of water utilizing the synchronously pumped Ti:Sapphire system**

$$V(\tau) = C\left[\int dt G_0^{(2)}(\tau - t) R(t)\right]$$

✓ **Autocorrelation signal**

OKE out-of-phase

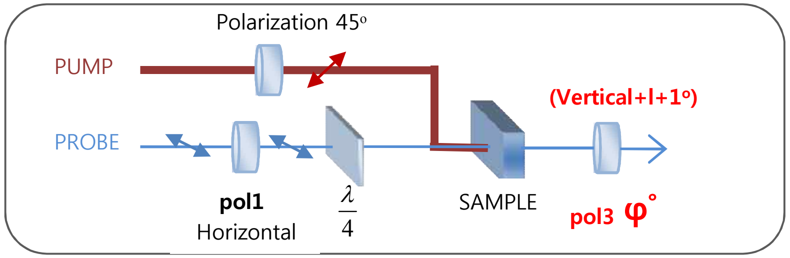


$$\underbrace{\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}}_{\text{pol3}}\underbrace{\begin{bmatrix}\cos 45^o & -\sin 45^o\\ \sin 45^o & \cos 45^o\end{bmatrix}\begin{bmatrix}e^{i\phi_x} & 0\\ 0 & e^{i\phi_y}\end{bmatrix}\begin{bmatrix}\cos 45^o & \sin 45^o\\ -\sin 45^o & \cos 45^o\end{bmatrix}}_{\text{SAMPLE}}\underbrace{\begin{bmatrix}1 & 0\\ 0 & i\end{bmatrix}}_{\frac{\lambda}{4}}$$

$$\underbrace{\begin{bmatrix}\cos\varphi & -\sin\varphi\\ \sin\varphi & \cos\varphi\end{bmatrix}\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}\begin{bmatrix}\cos\varphi & \sin\varphi\\ -\sin\varphi & \cos\varphi\end{bmatrix}}_{\text{pol1 }\varphi^\circ}\underbrace{\begin{bmatrix}1\\ 0\end{bmatrix}}_{\text{PROBE}} = \cos[\varphi]^2\sin[\frac{1}{2}(2\varphi+\phi_x-\phi_y)]^2$$

$$I = \langle E^2 \rangle = \varphi^2 + \varphi\Delta\phi + \frac{\Delta\phi^2}{4}$$

OKE in phase



$$\underbrace{\begin{bmatrix}\cos\varphi & -\sin\varphi\\ \sin\varphi & \cos\varphi\end{bmatrix}\begin{bmatrix}0 & 0\\ 0 & 1\end{bmatrix}\begin{bmatrix}\cos\varphi & \sin\varphi\\ -\sin\varphi & \cos\varphi\end{bmatrix}}_{\text{pol3 }\varphi^\circ}$$

$$\underbrace{\begin{bmatrix}\cos 45^o & -\sin 45^o\\ \sin 45^o & \cos 45^o\end{bmatrix}\begin{bmatrix}e^{i\phi_x} & 0\\ 0 & e^{i\phi_y}\end{bmatrix}\begin{bmatrix}\cos 45^o & \sin 45^o\\ -\sin 45^o & \cos 45^o\end{bmatrix}}_{\text{SAMPLE}}$$

$$\underbrace{\begin{bmatrix}1 & 0\\ 0 & i\end{bmatrix}}_{\frac{\lambda}{4}}\underbrace{\begin{bmatrix}1 & 0\\ 0 & 0\end{bmatrix}}_{\text{pol1}}\underbrace{\begin{bmatrix}1\\ 0\end{bmatrix}}_{\text{PROBE}} = \frac{1}{4}(2-\cos[2\varphi+\phi_x-\phi_y]-\cos[2\varphi-\phi_x+\phi_y])$$

$$I = \langle E^2 \rangle = \frac{1}{2} - \frac{1}{2}\cos\Delta\phi\cos 2\varphi$$

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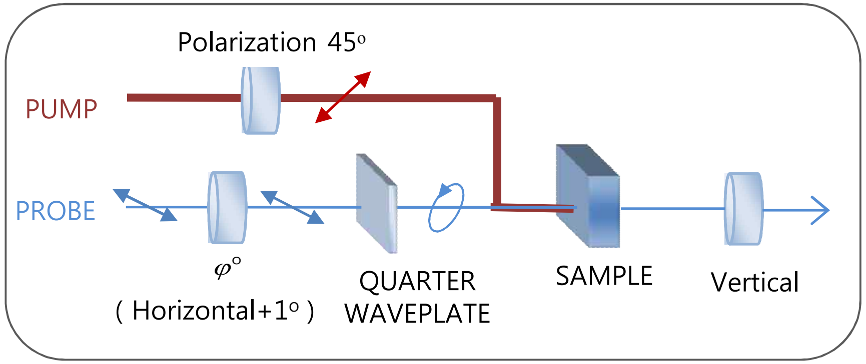
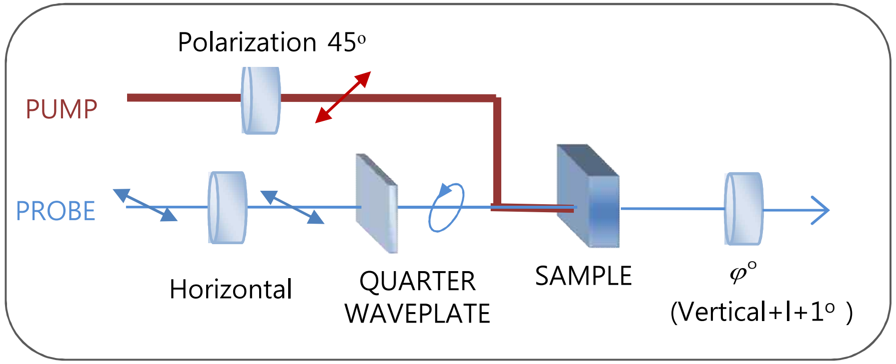
✓ out-of-phase

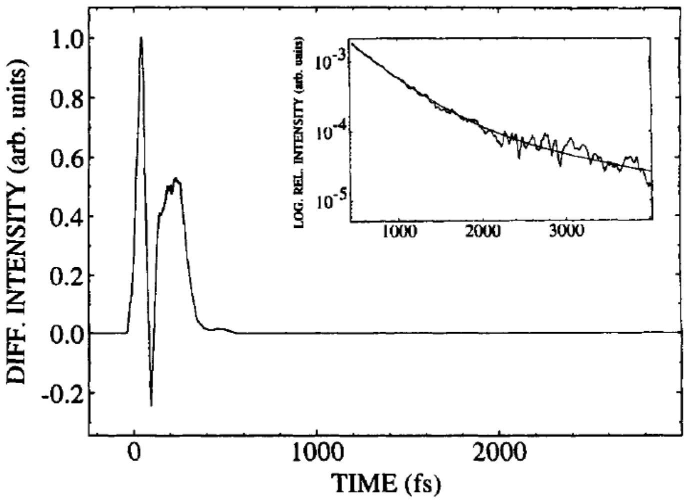


$$I = \langle E^2 \rangle = \varphi^2 + \varphi\Delta\phi + \frac{\Delta\phi^2}{4}$$

✓ in-phase



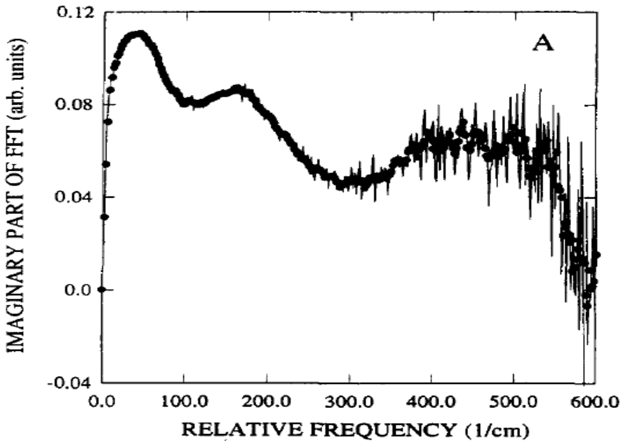
$$I = \langle E^2 \rangle = \frac{1}{2} - \frac{1}{2}\cos\Delta\phi\cos 2\varphi$$



✓ **In-phase heterodyne-detected results utilizing the amplified CPM system**

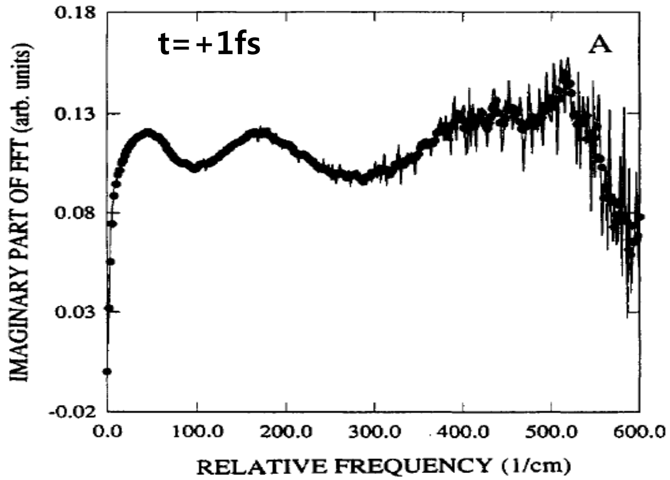
✓ **The absolute magnitude of the in-phase response was more than 1 order of magnitude less than out-of-phase signal under the same condition**

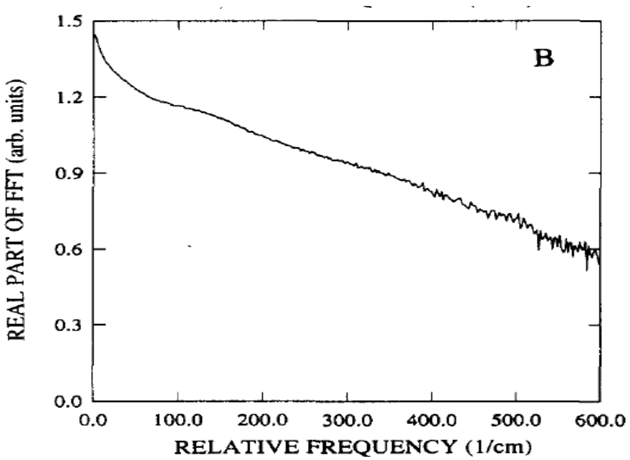
Frequency domain OKE of water with an optimal t=0 position position obtained from the Fourier transform of the out-of-phase heterodyne response from fig2



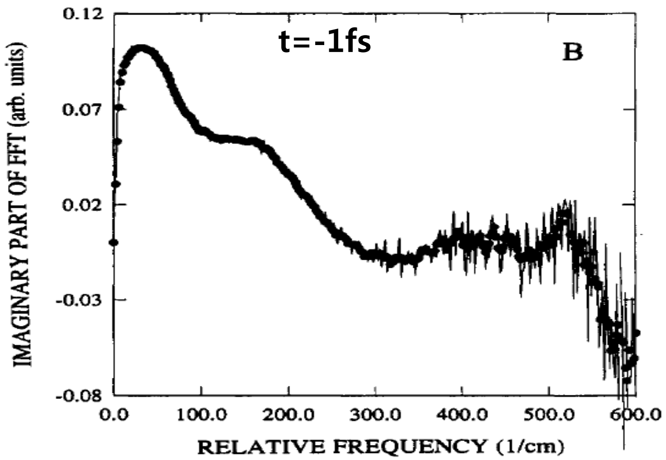
(A) Imaginary part of $\chi^{(3)}$

$$F(V_L(\tau)) = F(G_0^{(2)}(\tau))F(R(t))$$





(B) Real part of $\chi^{(3)}$



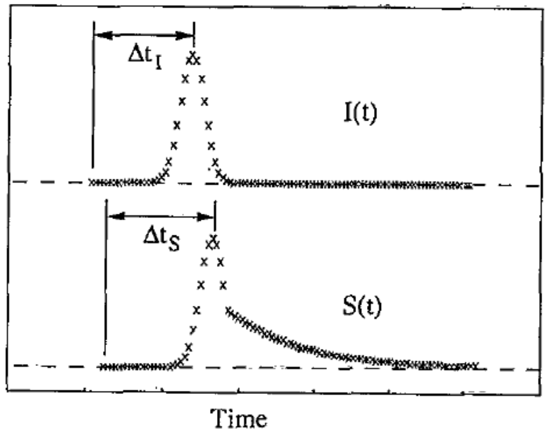


FIG. 1. Model laser autocorrelation function (top) and synthetic raw signal (bottom). The crosses represent individual data points. Δt_I and Δt_S denote the time gaps between the first and the maximum data points. Since the absolute zero time cannot be maintained for measurements of the laser autocorrelation function and the raw signal, Δt_I and Δt_S should be considered to be independent quantities. Taking Δt_S as an adjustable parameter (we could equally well take Δt_I), the spectral density $\mathrm{Im}[\chi(\omega)]$ can be calculated (see the text for details).

$$S(t) \cong \Delta\sigma I(t) + \int_{-\infty}^{\infty} d\tau\, I(t-\tau)\Delta\chi(\tau)$$

$$\mathrm{Im}[\Delta\chi(\omega)] = \mathrm{Im}[S(\omega)/I(\omega)],$$

$$\mathrm{Re}[\Delta\chi(\omega)] = \mathrm{Re}[S(\omega)/I(\omega)] - \Delta\sigma.$$

$$S(\omega) = S'(\omega)\exp(i\omega\Delta t_s)$$

$$I(\omega) = I'(\omega)\exp(i\omega\Delta t_I)$$

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