Antiresonant-ring Kerr spectroscopy

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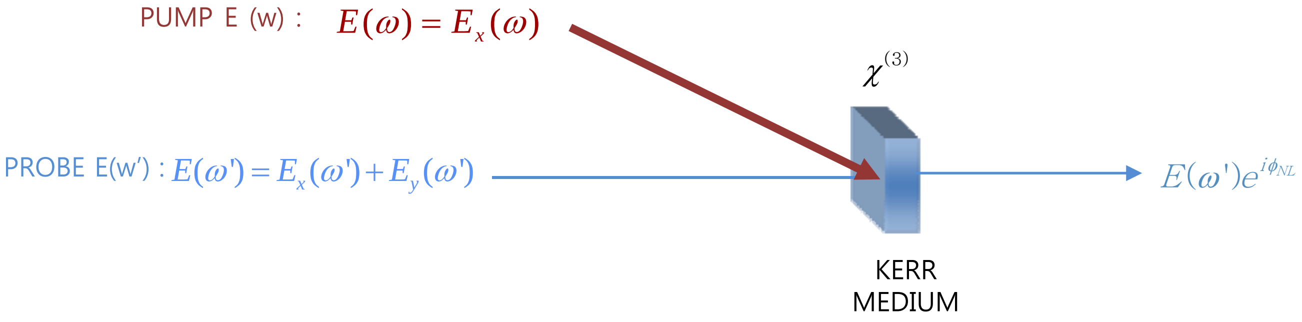
Introduction

- **Generally Optical Kerr Effect**



$$P_x^{(3)}(\omega') = \chi_{1111}^{(3)}(\omega' = \omega' + \omega - \omega)E_1(\omega')E_1(\omega)E^*{}_1(\omega) = (\chi_{1122}^{(3)} + \chi_{1212}^{(3)} + \chi_{1221}^{(3)})(\omega' = \omega' + \omega - \omega)E_x(\omega')E(\omega)E^*(\omega)$$

$$P_y^{(3)}(\omega') = \chi_{1122}^{(3)}(\omega' = \omega' + \omega - \omega)E_y(\omega')E(\omega)E^*(\omega)$$

$$\boxed{\delta\chi(\omega') = \Delta\chi_{xx} - \Delta\chi_{yy} = (\chi_{1212} + \chi_{1221})\left|E_0(\omega)\right|^2}$$

in isotropic medium

- **Mode Selective OKE**

measured $\chi_{1111}^{(3)}, \chi_{1122}^{(3)}$

Experimental setup

- Ti:sapphire laser 50fs 800nm
- pump beam : travers optical delay line
- probe beam : polarization 45^{o}
  one probe pulse arrive at the sample several picosecond or more before the other
- dual-ring interferometer two polarizing beam cube (**PBC**)
- blue denote polarizations before sample
- red denote polarizations after sample

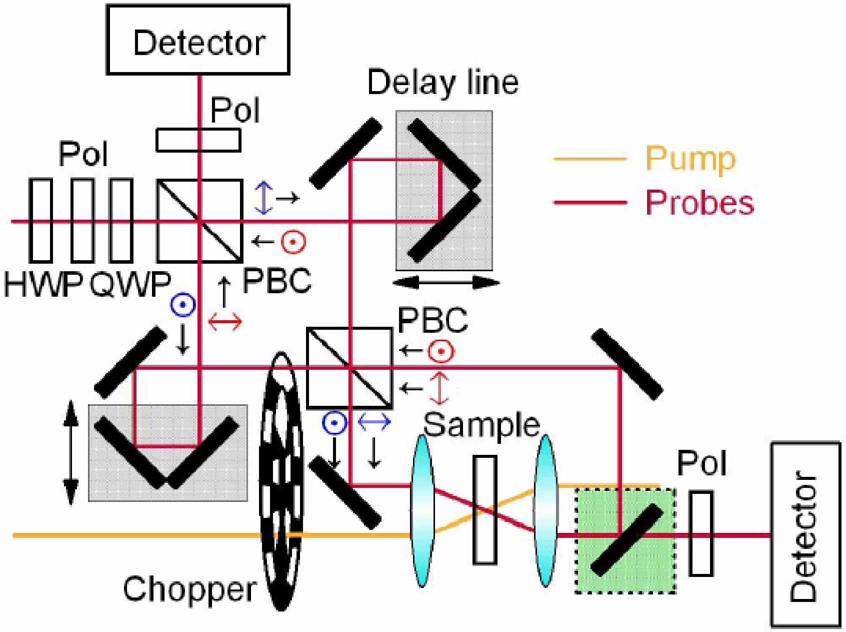
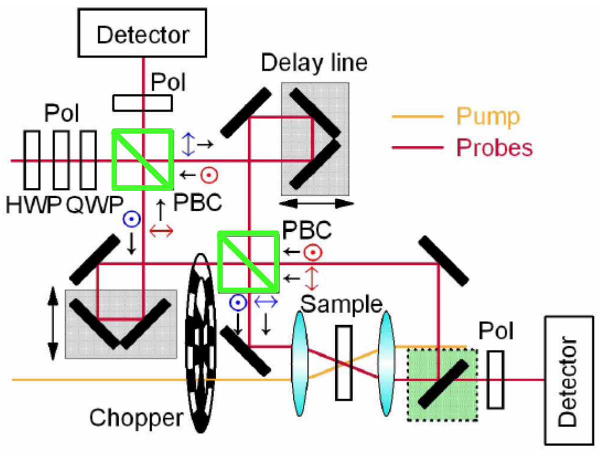


Fig. 1. Experimental setup for ARKS. HWP = half-wave plate, Pol = polarizer, QWP = quarter-wave plate, PBC = polarizing beam cube, ⊙ = vertical polarization, ↔ = horizontal polarization. Blue denotes polarizations before the sample and red polarizations after the sample. The mirror in the green block can be removed to measure the depolarized OKE response.

Experimental setup



PBC extinction ratio :
 transmitted direction horizontal:vertical=200:1
 reflected direction vertical:horizontal=lower

Vertical polarized light can be ignored, but there are significant reflections of the horizontal polarization component

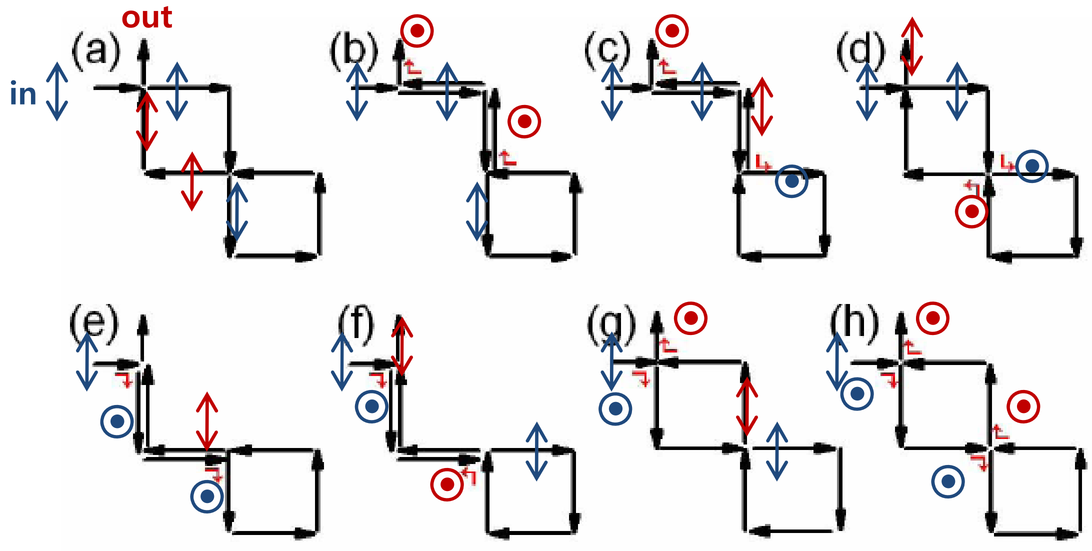


Fig. 2. Pathways through which horizontally-polarized light that enters the ARKS apparatus can exit in the direction in which the signal is detected. Reflections of the predominantly transmitted horizontal polarization are indicated with red arrows.

(a) desired path

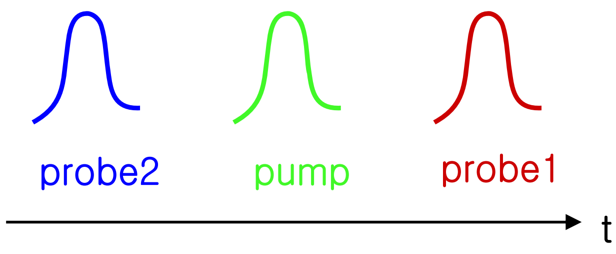
(b)(c)(e)(f) change the path length of the probe beam (arrives at the detector at a significantly different time from the signal

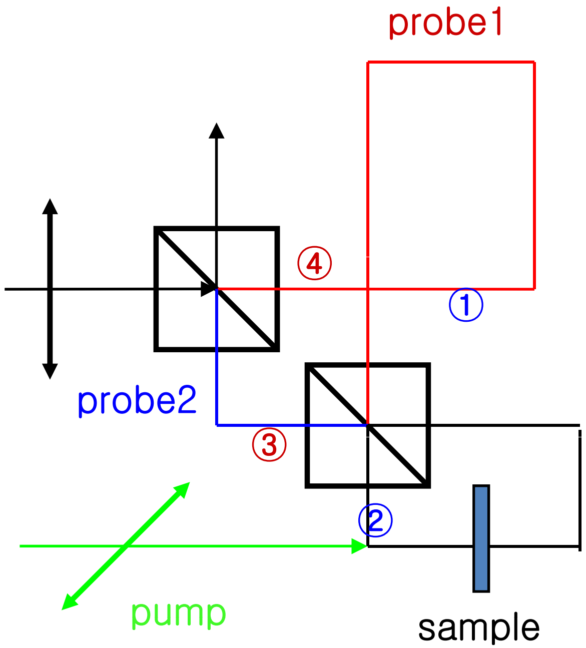
(d)(g) passes through the sample in the wrong

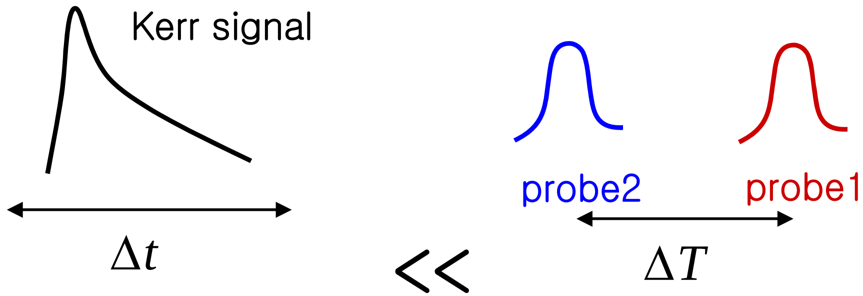
(h) very weak

Experimental setup

- polarization : **pump 45°**
- one probe beam (reflected beam at first PBS, polarization **y**) is set to arrive at the sample before the other (polarization **x**)
- time between the probe beams >> pump and probes time delay (pump does not affect the earlier probe, so only later probe phase change due to pump)







$$P_x = \chi_{xxxx} + \chi_{xxyy}$$

③ ④

Results

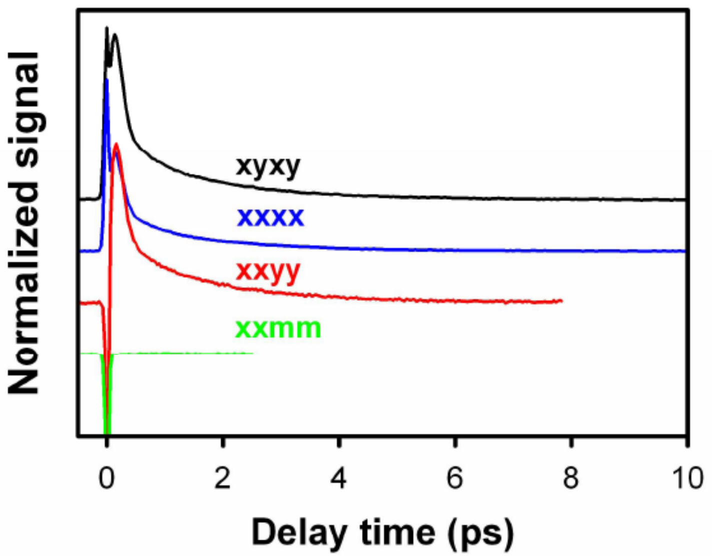


Fig3. Normalized ARKS data for CS_2 under difference polarization condition. The data have been offset vertically for clarity. xxmm (isotropic response))

- depolarized response dominance:
  similarities, xyxy, xxxx, xxyy tensor elements
- lack of isotropic response

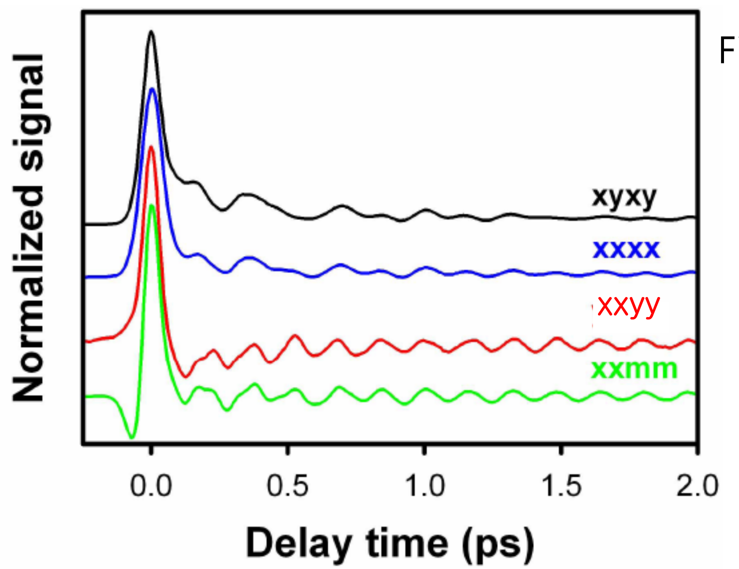


Fig4. Normalized ARKS data for S_2Cl_2 under difference polarization conditions. The data have been offset vertically for clarity.

- sililar, xxyy and xxmm

Results

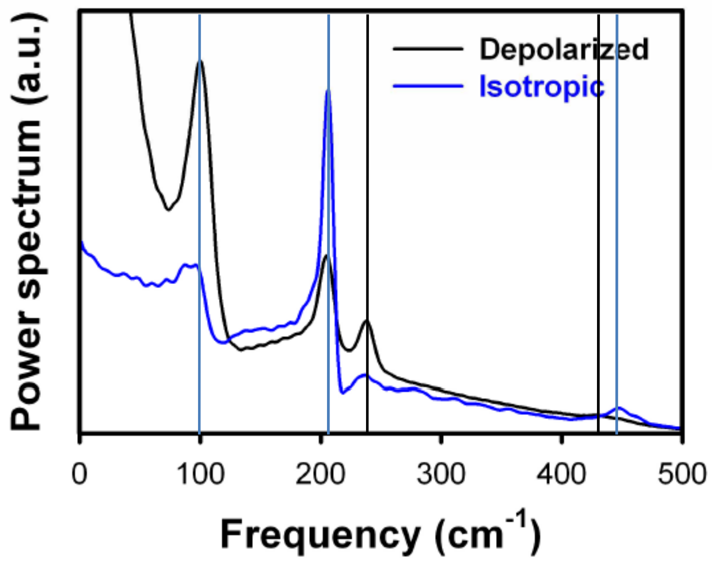


Fig5. Depolarized and isotropic OKE power spectra for S_2Cl_2. The two spectra are not to scale.

Depolarized response : collective molecular response, librational, collective scattering from depolarized intermolecular modes_ _ induced by the pump pulse

Isotropic response : intramolecular and intermolacular modes with the appropriate symmetry

102cm^{-1}
v_4(torsion)

205cm^{-1}
v_3(S-S-Cl symmetric angle deformation)

446cm^{-1}
v_2(S-Cl symmetric strength)

238cm^{-1}
v_6(S-S-Cl asymmetric angle deformation)

434cm^{-1}
v_5(S-Cl asymmetric strength)

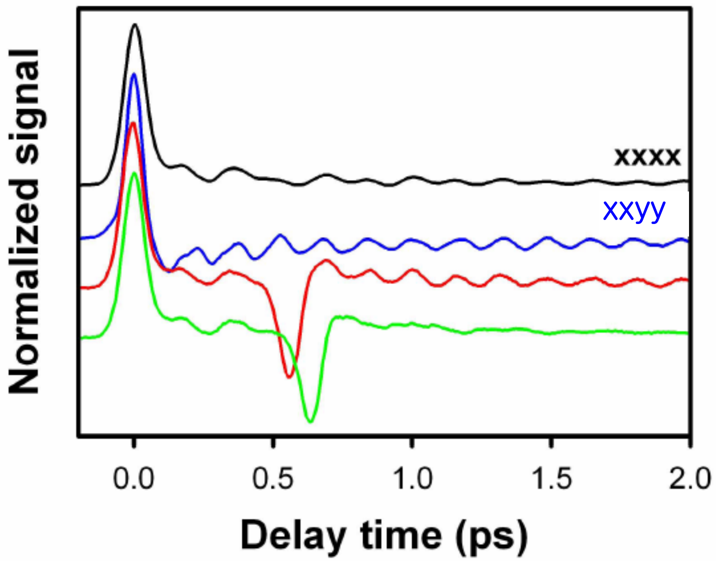


Fig6. Normalized S_2Cl_2 ARKS data for the xxxx and yyxx tensor elements, as well as scans in which these tensor elements are optically subtracted with different time shifts to enhance or suppress the contribution of the v_3 mode.

- xxxx, xxyy : one probe arrive before the pump pulse
- lower two traces : two probe arrive after the pump pulse
- v_2 vibration (red trace)
- suppress it virtually completely (green trace)