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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
34100 TRIESTE (ITALY) - P.O.B. 586 - MIRAMARE - STRADA COSTIERA 11 - TELEPHONES: 224281/2/3/4/5/6
CABLE: CENTRATOM - TELEX 460392-I

SMR/100 - 58

WINTER COLLEGE ON LASERS, ATOMIC AND MOLECULAR PHYSICS

(24 January - 25 March 1983)

CARS Spectroscopy

Part I

General Introduction

G. MAROWSKY

Max-Planck-Institut für
Biophysikalische Chemie
Göttingen
Fed. Rep. Germany

Coherent

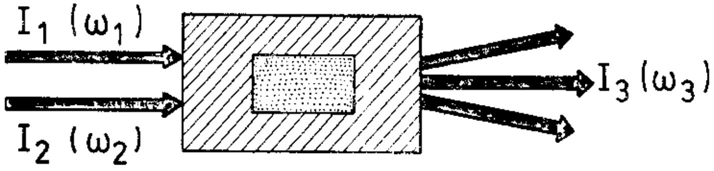
Antistokes

Raman

Scattering

1963: Maker und Terhune

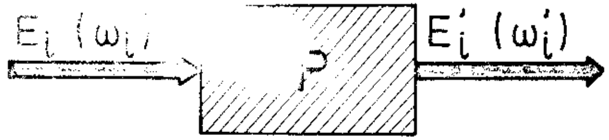
Benzene: C_6H_6



$$\omega_1 - \omega_2 = \Delta\omega_{Raman} = 992\,cm^{-1}$$

1/2

Interaction Light - Matter:



$$P = P_L + P_{NL}$$

$$= \{\sigma_i, \omega_i, N, \tau_i\}$$

Linear Polarisation:

$$\vec{P_L} = \alpha\vec{E} = \varepsilon_0 \chi^{(1)}\vec{E}$$

$$\vec{D} = \varepsilon_0\vec{E} + \vec{P}$$

$$\varepsilon\varepsilon_0 = \varepsilon_0(1+\chi^{(1)}) = \varepsilon_0 n^2$$

Symbolic Ansatz for P:

$$P\{E_i(\omega_i)\} =$$

$$\varepsilon_0\{\chi^{(1)}E_1 + \chi^{(2)}E_1E_2 + \chi^{(3)}E_1E_2E_3 + \dots\}$$

Special Cases,
using one, two, or three
different frequencies
ω_i.

$$P\left\{E_i(\omega_i)\right\} =$$
$$\varepsilon_0\left\{\chi^{(1)}E_1 + \chi^{(2)}E_1E_2 + \chi^{(3)}E_1E_2E_3 + \cdots\right\}$$

$$\chi^{(3)}(\omega_3;\omega_a,\omega_b,\omega_c)$$
$$\omega_3 = \omega_a + \omega_b + \omega_c$$

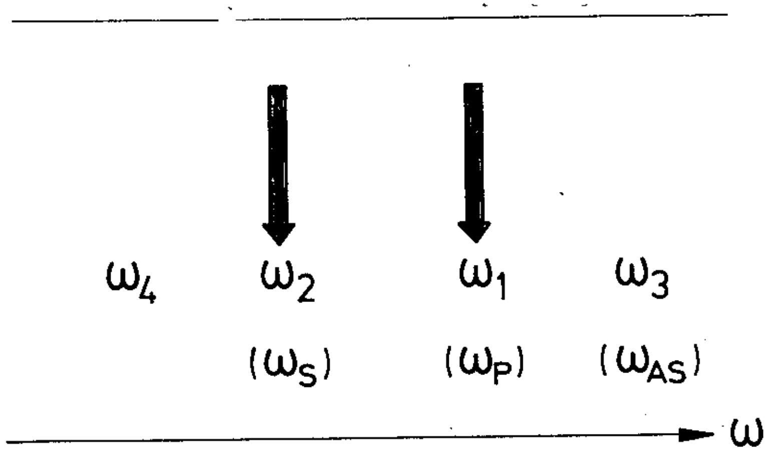
1.) $\omega_3 = 3\omega$

2.) $\chi^{(3)} = \chi^{(3)}(\omega_2;\omega_1,-\omega_1,\omega_2)$
"RIKES"

3.) $\chi^{(3)} = \chi^{(3)}(\omega_3;\omega_1,\omega_1,\pm\omega_2)$
-: "CARS" - Type
+: 2-Photon- Type

4.) $\chi^{(3)} = \chi^{(3)}(\omega_3;\omega_0,\omega_1,-\omega_2)$
"4-WMP"

Simple Explanation: "Sidebands"



$$\omega_1 - \omega_2 = \Delta\omega_R$$

$$\omega_2 - \boxed{\omega_3}$$
CARS

$$\boxed{\omega_4} - \omega_1$$
CSRS

$\chi^{(3)}$ Philosophie

$$E_1 = \tilde{E}_1 \cdot e^{i(k_1 x - \omega_1 t)}$$

$$E_2 = \tilde{E}_2 \cdot e^{i(k_2 x - \omega_2 t)}$$

$P_{NL}(\omega_3) = ?$

$$\omega_3 = \omega_{AS} = 2\omega_1 - \omega_2$$

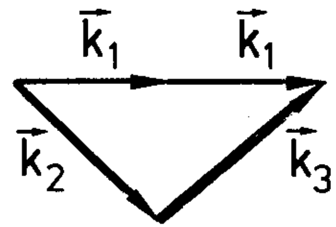
$\boxed{E_1 \times E_1 \times E_2^*}$ CARS

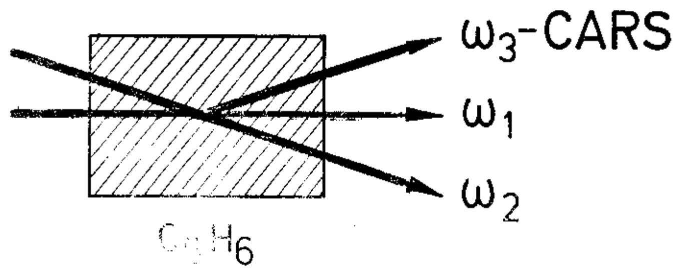
$\boxed{E_1 \times E_1^* \times E_2^*}$ Raman

$$P_{NL}(\omega_3) \longrightarrow E_3(\omega_3, \vec{k}_3)$$

$\omega_3 = 2\omega_1 - \omega_2$ "Energy"

$\vec{k}_3 = 2\vec{k}_1 - \vec{k}_2$ "Momentum"





Structure of $\chi^{(3)}$

$$\chi^{(3)} = \chi_E^{(3)} + \chi_{NR}^{(3)} + \chi_{Raman}^{(3)}$$

$$\chi_{Raman}^{(3)} = \tilde{\chi}_R^{(3)} \cdot \frac{\Gamma_R}{(\Delta\omega_R - (\omega_1 - \omega_2) + i\Gamma_R)}$$

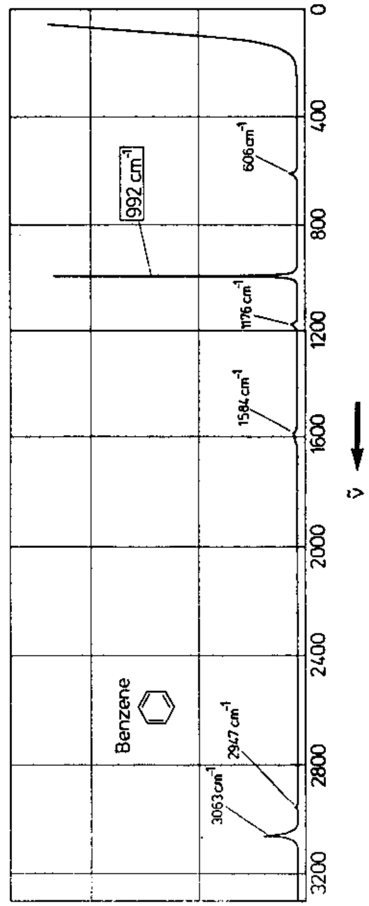
$$= \chi_{Raman}^{(3)'} + i\chi_{Raman}^{(3)''}$$

$$\tilde{\chi}_R^{(3)} \sim \frac{N\pi c^4}{\hbar\omega^4\Gamma_R} \cdot \left(\frac{d\sigma}{d\Omega}\right)_{Raman,\ spontan}$$

$$P_{AS} \sim \left| \chi^{(3)} \right|^2$$

$$\text{d.h.} \sim N^2, \ \left| \tilde{\chi}_R^{(3)} \right|^2, \ \left(\frac{d\sigma}{d\Omega}\right)^2$$

Raman Data on Benzene



ε: *Efficiency*

$\varepsilon = P_{AS} / P_2(\omega_2)$

$$\varepsilon \sim \frac{\left|N \cdot \chi^{(3)}\right|^2}{\lambda^2} \times P_1(\omega_1)^2 \times \left[\frac{\sin\left(\frac{\Delta kz}{2}\right)}{\left(\frac{\Delta kz}{2}\right)}\right]^2 \times \left(\frac{z}{2}\right)^2$$

$\Delta k = 2k_1 - k_2 - k_3$

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1. $\Delta k = 0$: $\varepsilon \sim z^2$

   $z \longrightarrow$ *Beam Overlapp*

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2. $\Delta k \neq 0$ (z.B. kollinear *Propagation*)

   $z \rightarrow L_c = \pi / \Delta k$

   $$\sin\left(\frac{\Delta kz}{2}\right) = 1$$

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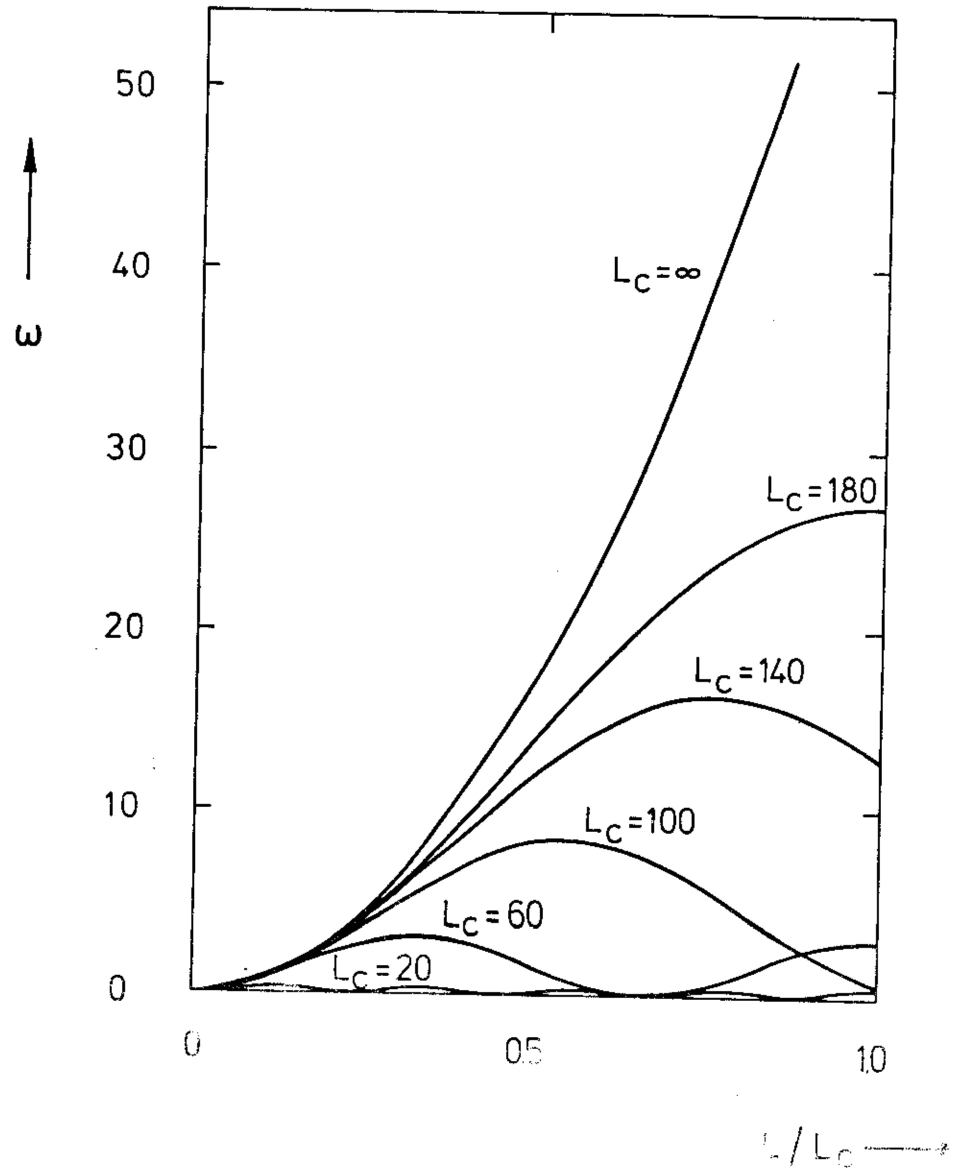
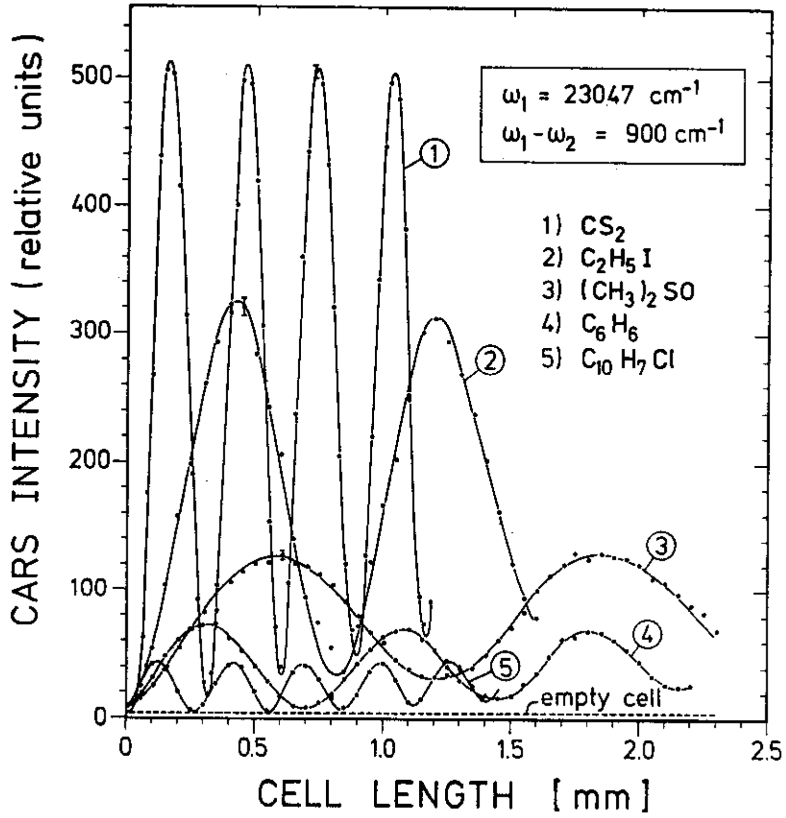
3. Gaußian *Beams*:

   $P_1^2 \longrightarrow \left(\frac{P_1}{A}\right)^2$; $A = \pi r^2$

   $L_c \longrightarrow b = 2\pi \cdot \left(\frac{r^2}{\lambda}\right) \longrightarrow \varepsilon \sim P_1^2(\omega_1)$

   b: konfokal Parameter





S.Saikan, G.Marowsky: Opt. Commun. 26 (1978), 466

psec – CARS