

# My Early Contributions to Nonlinear Optics

Invited Lecture by Ray Chiao  
at the International Conference  
“Nonlinear Optics East-West Reunion” in  
Suzdal, Russia, Sept. 21, 2011.

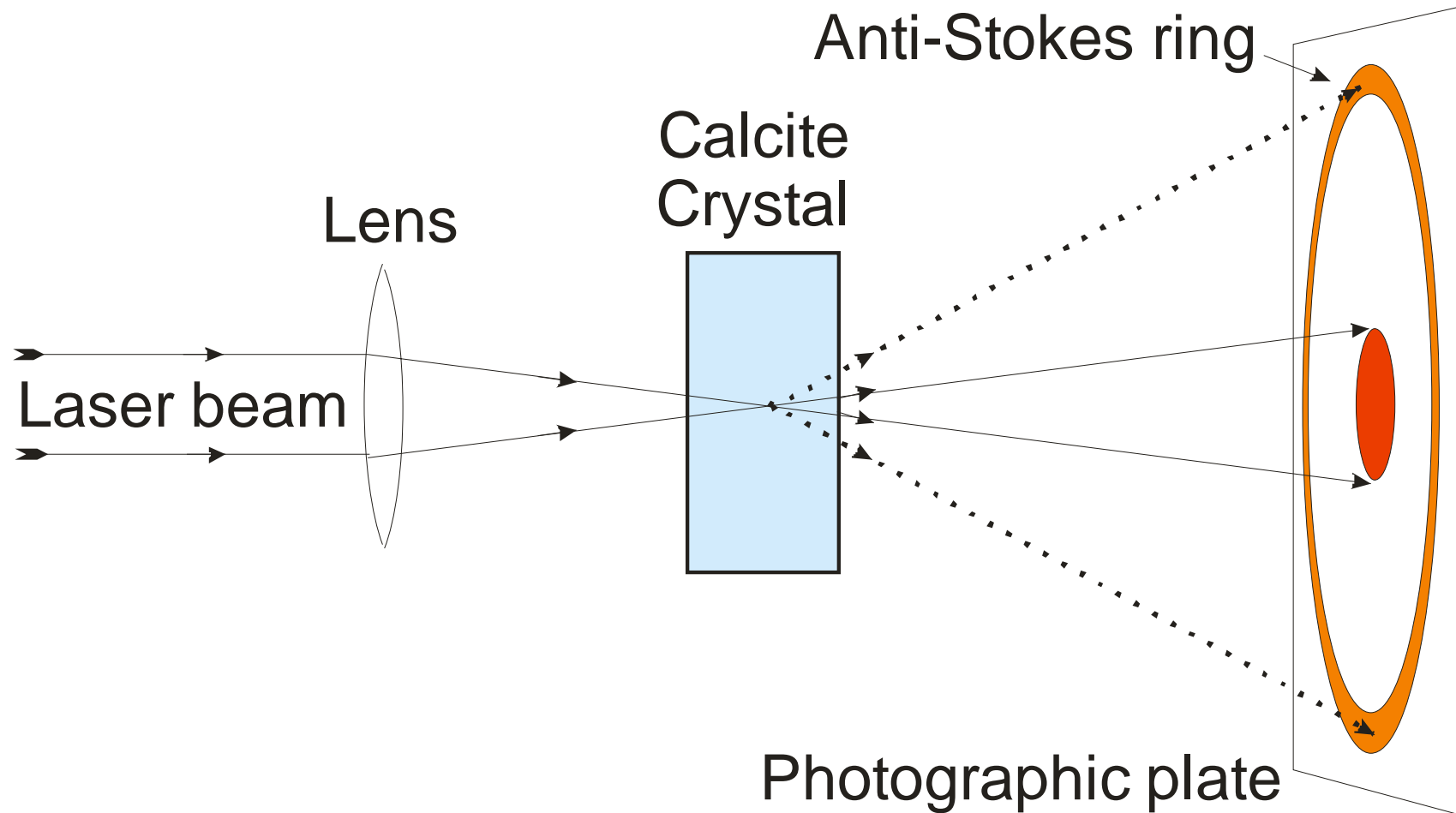
# Outline

- Stimulated Raman Scattering in Calcite (Chiao & Stoicheff, 1964)
- Stimulated Brillouin Scattering (Chiao, Townes, Stoicheff, 1964)
- Self-Trapping of Optical Beams (Chiao, Garmire, Townes, 1964)
- Weak-Wave Retardation and Degenerate Four-Wave Parametric Gain (Chiao, Kelley, Garmire 1966; Carman, Chiao, Kelley, 1966).

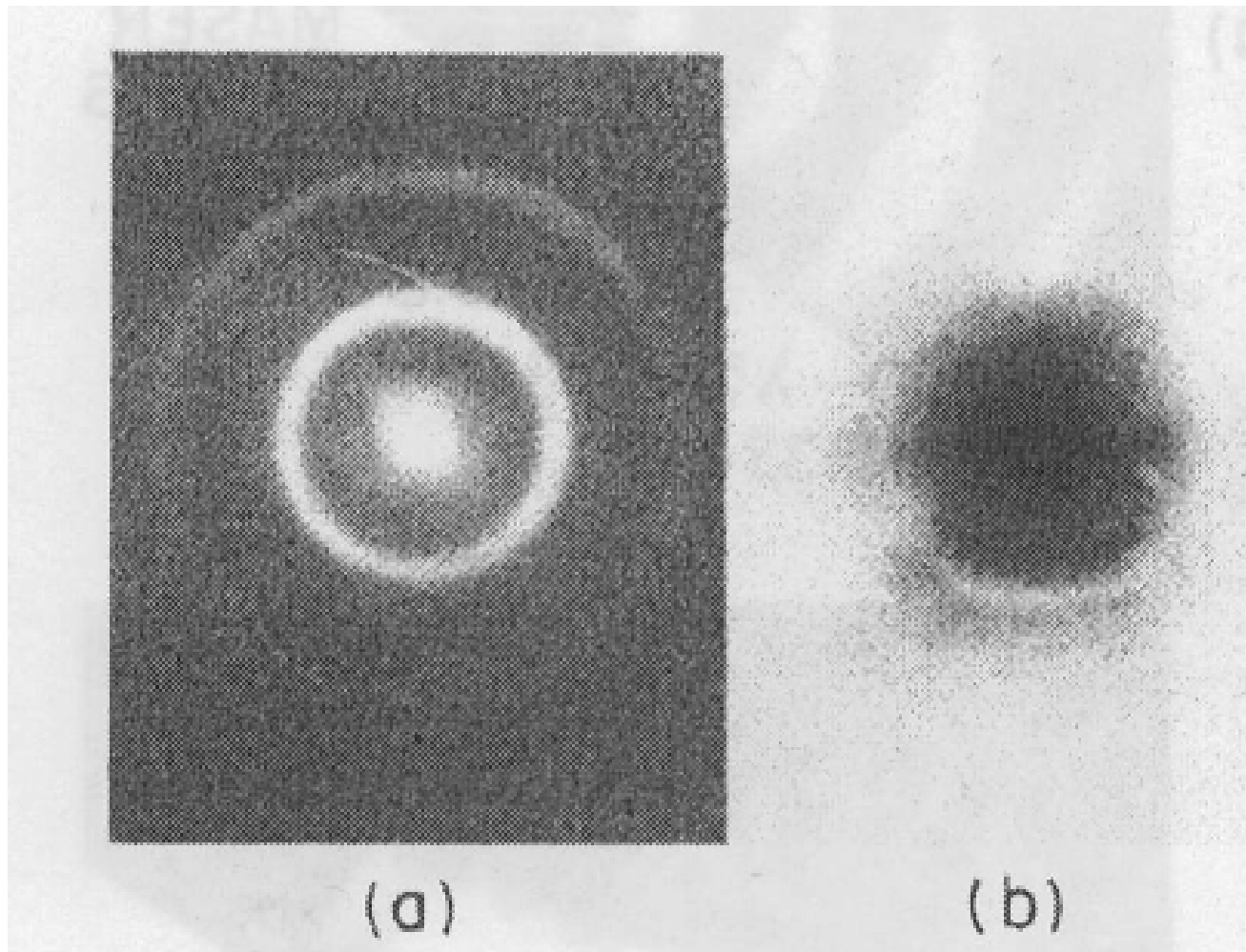
# Angular Dependence of Stimulated Raman Radiation in Calcite

R. Chiao and B. P. Stoicheff,  
Phys. Rev. Letters 12, 290 (1964)

# Stimulated Raman Scattering in a Calcite Crystal

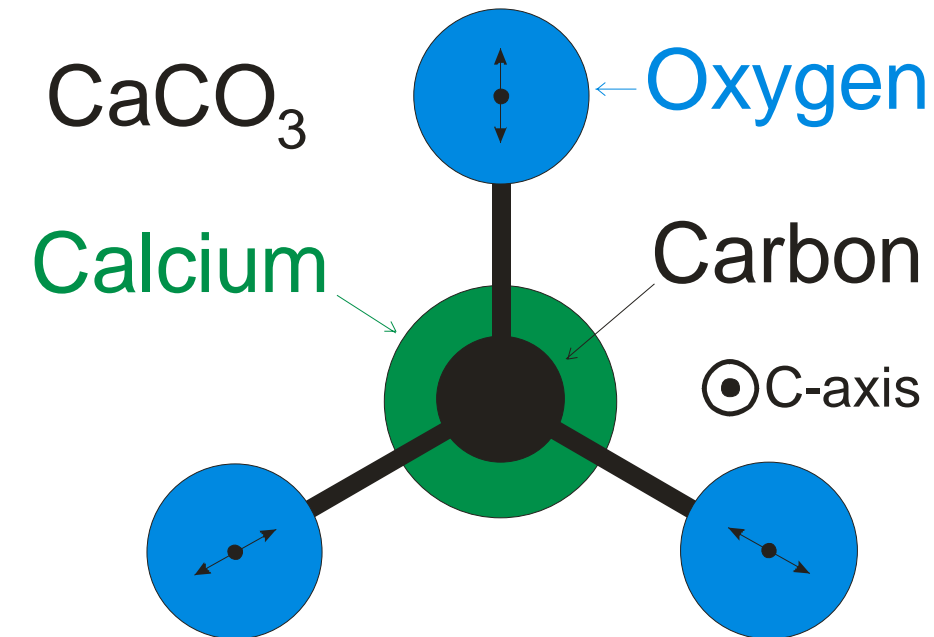


# Anti-Stokes (a) and Stokes (b) Rings in Stimulated Raman Scattering in Calcite



# Calcite crystal structure

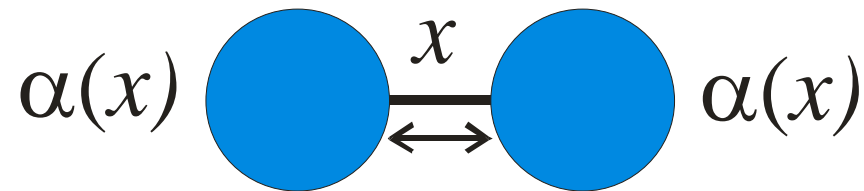
- Calcite is composed of stacks of the calcium carbonate molecule, which has the following structure:



“Symmetric stretch”  
is the radial breathing mode  
of the calcium carbonate molecule  
at a resonance frequency of  $1085.6 \text{ cm}^{-1}$

# Herzberg's Raman-Effect Model

- Consider a diatomic molecule whose polarizability  $\alpha(x)$  is a function of its bond “stretch length”  $x$ .



- Then a simple-harmonic-oscillator variation in  $x$  will result in a simple-harmonic-oscillator variation in the polarizability  $\alpha(x)$  of the molecule. This will create *sidebands* in the laser light because

$$\alpha(x) \approx \alpha(0) + \frac{d\alpha}{dx}x + \dots$$

# Garmire, Pandarese & Townes: Simple harmonic oscillator model for SRS

- The energy  $U(x)$  of a diatomic molecule placed in an electric field in Herzberg's model is

$$U(x) = \frac{1}{2} \alpha(x) E^2$$

- Force stretching the molecule in a laser beam is

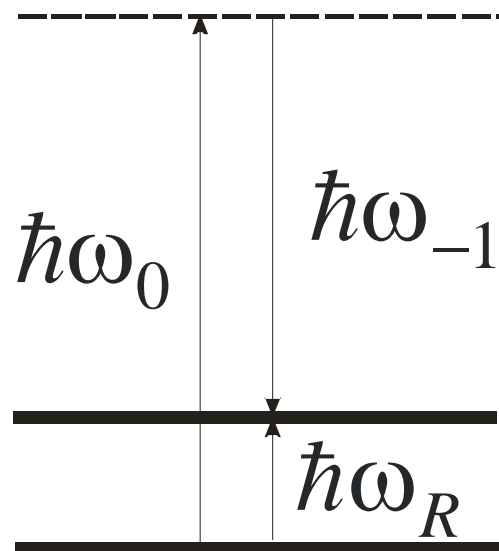
$$F = -\frac{dU}{dx} = -\frac{d\alpha}{dx} E^2$$

- Force due to the *beating* between a laser  $\mathbf{E}_0$  and the first Stokes wave  $\mathbf{E}_{-1}$  will drive the molecule *resonantly*.



# First Stokes Radiation in Stimulated Raman Scattering

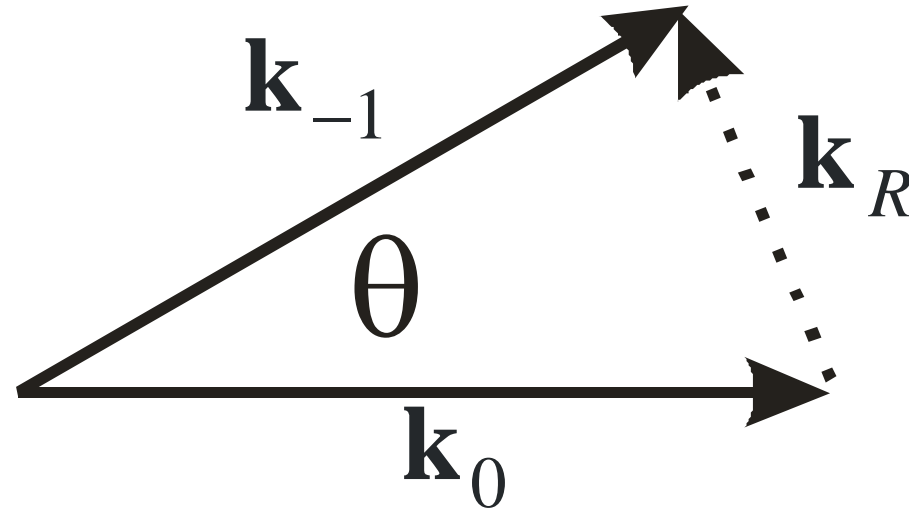
- Energy level diagram for Raman effect



$$\hbar\omega_0 - \hbar\omega_{-1} = \hbar\omega_R$$

$$\text{or, } \omega_0 - \omega_{-1} = \omega_R$$

# Phase-matching Condition for 1st Stokes

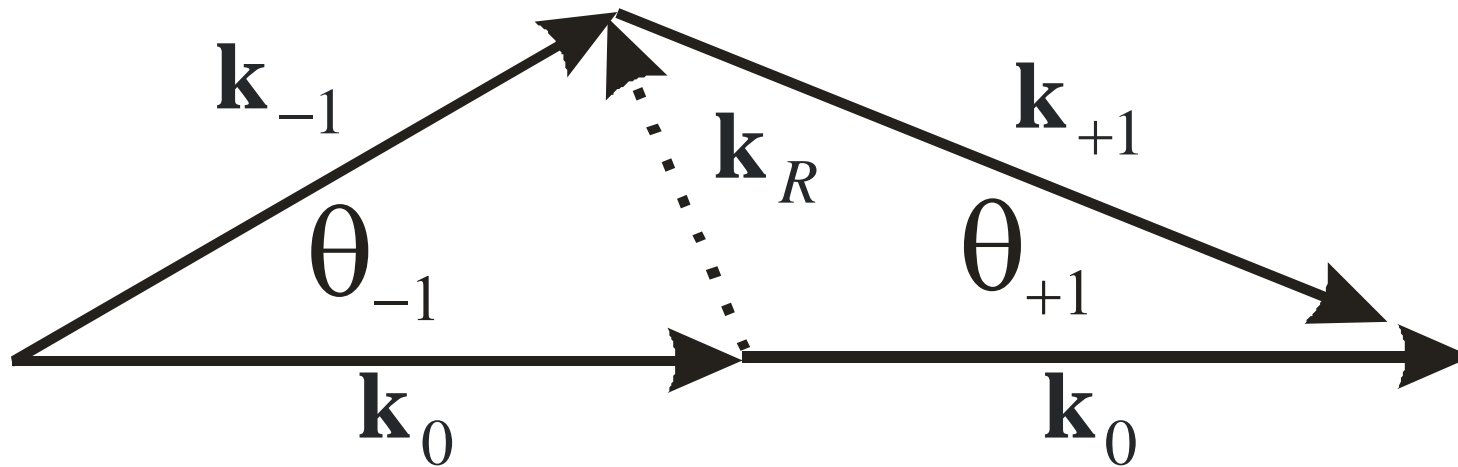


$$\omega_0 - \omega_{-1} = \omega_R$$

$$\mathbf{k}_0 - \mathbf{k}_{-1} = \mathbf{k}_R$$

Since  $\omega_R$  is independent of  $\mathbf{k}_R$ , stimulated Raman gain occurs for *all* scattering angles  $\theta$ .

# Phase-matching Condition for 1st Anti-Stokes Ring

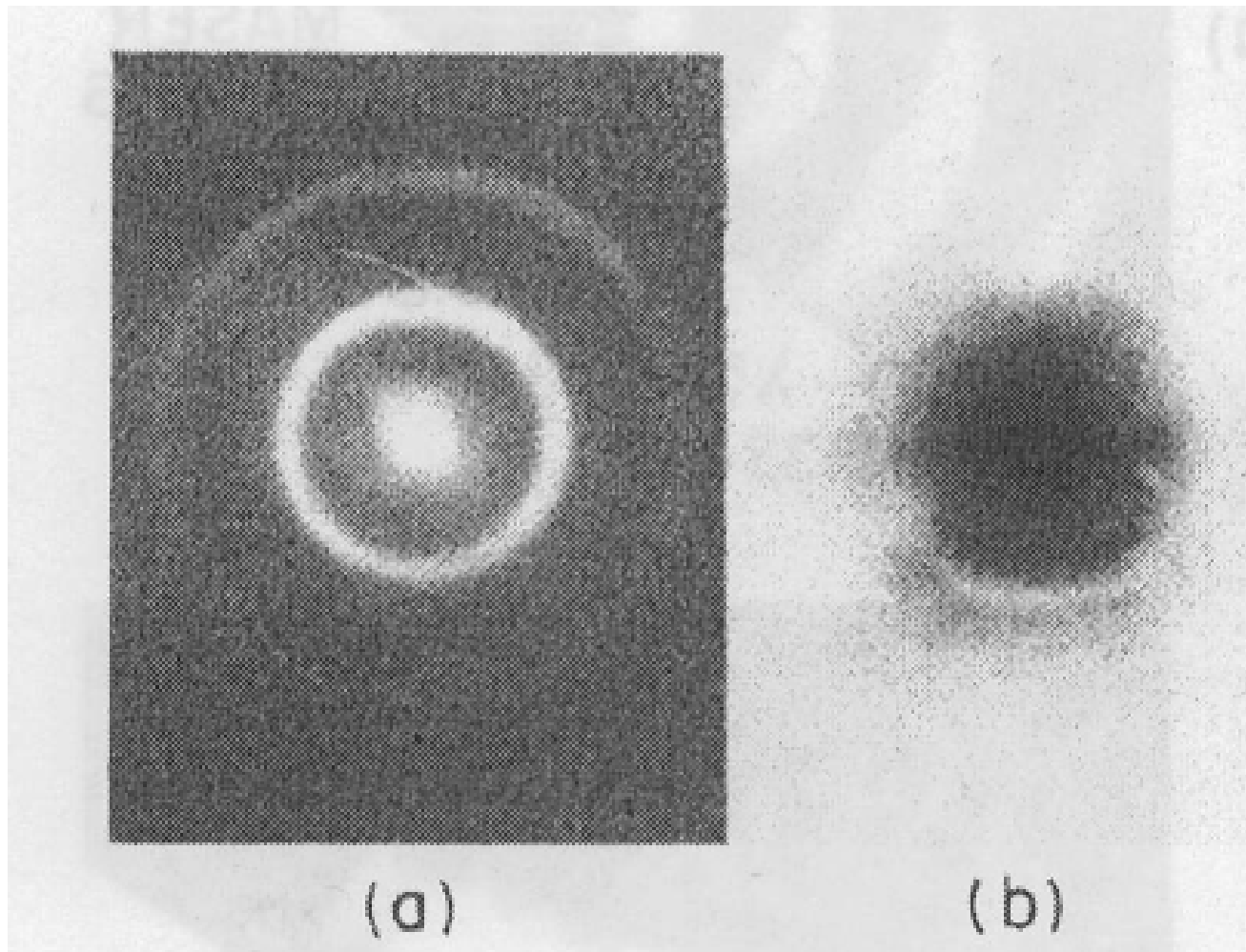


$$\omega_0 - \omega_{-1} = \omega_R = \omega_{+1} - \omega_0$$

$$k_0 - k_{-1} = k_R = k_{+1} - k_0$$

This is a *two-step* process, not a *four-wave mixing* process.

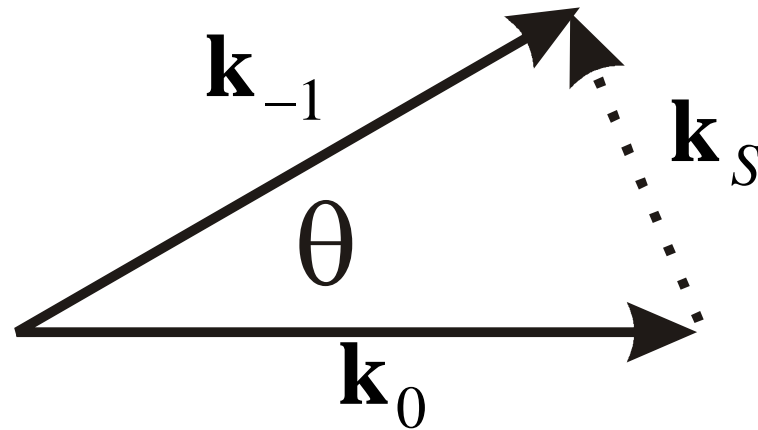
# Anti-Stokes (a) and Stokes (b) Rings in Stimulated Raman Scattering in Calcite



# Stimulated Brillouin Scattering

R. Y. Chiao, C. H. Townes, and  
B. P. Stoicheff, Phys. Rev. Letters  
12, 592 (1964)

# Brillouin Scattering: Stokes case

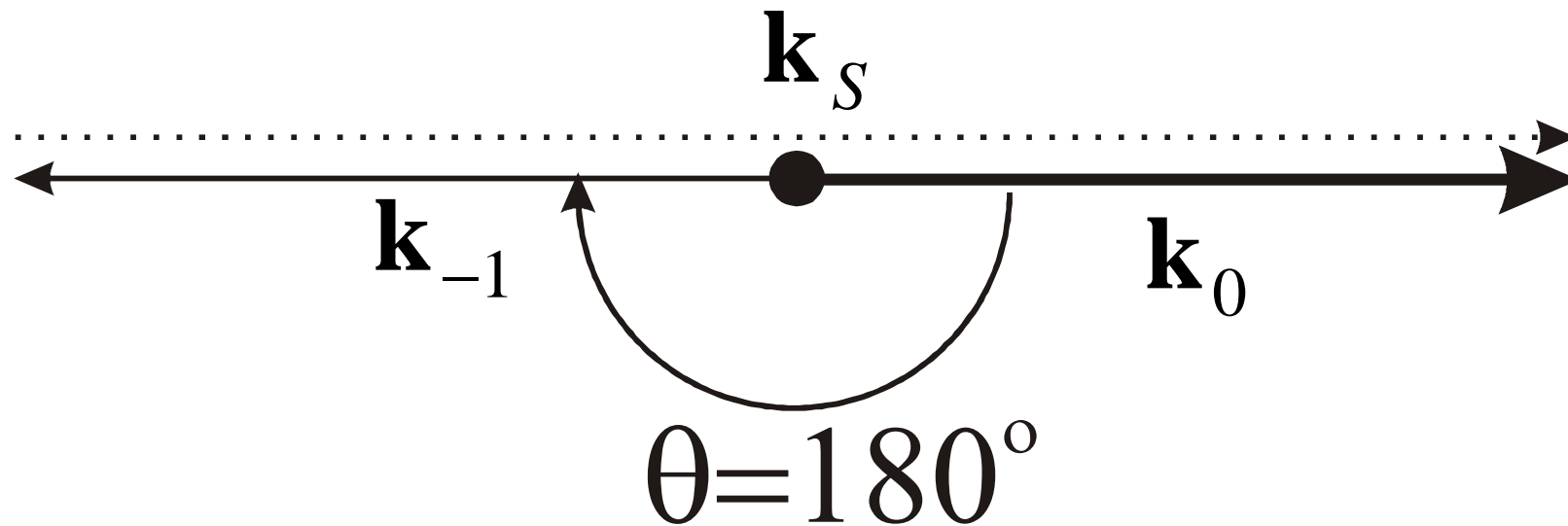


$$\omega_0 - \omega_{-1} = \omega_S$$

$$\mathbf{k}_0 - \mathbf{k}_{-1} = \mathbf{k}_S$$

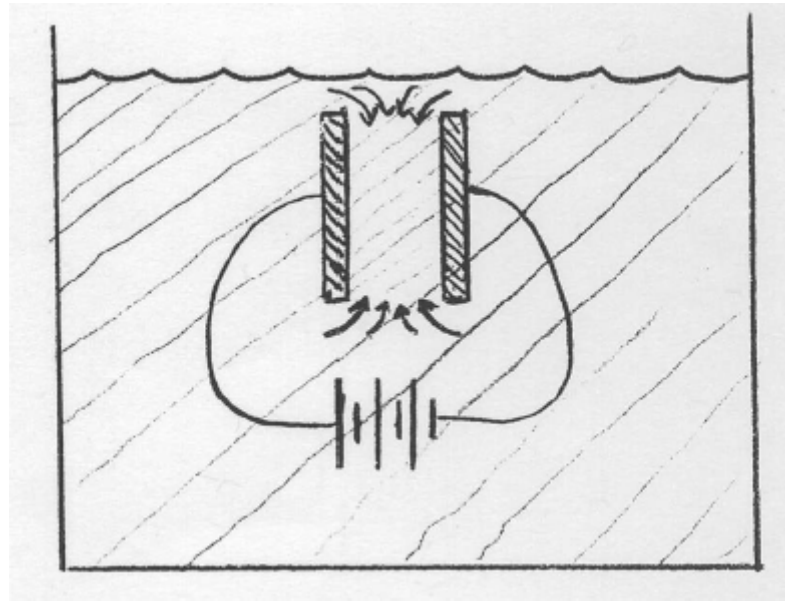
$$\omega_S = 2n\omega_0 \left( \frac{v_S}{c} \right) \sin \left( \frac{\theta}{2} \right)$$

# Stimulated Brillouin Back-Scattering



$$\omega_0 - \omega_{-1} = \omega_s = 2n\omega_0 \left( \frac{v_s}{c} \right)$$

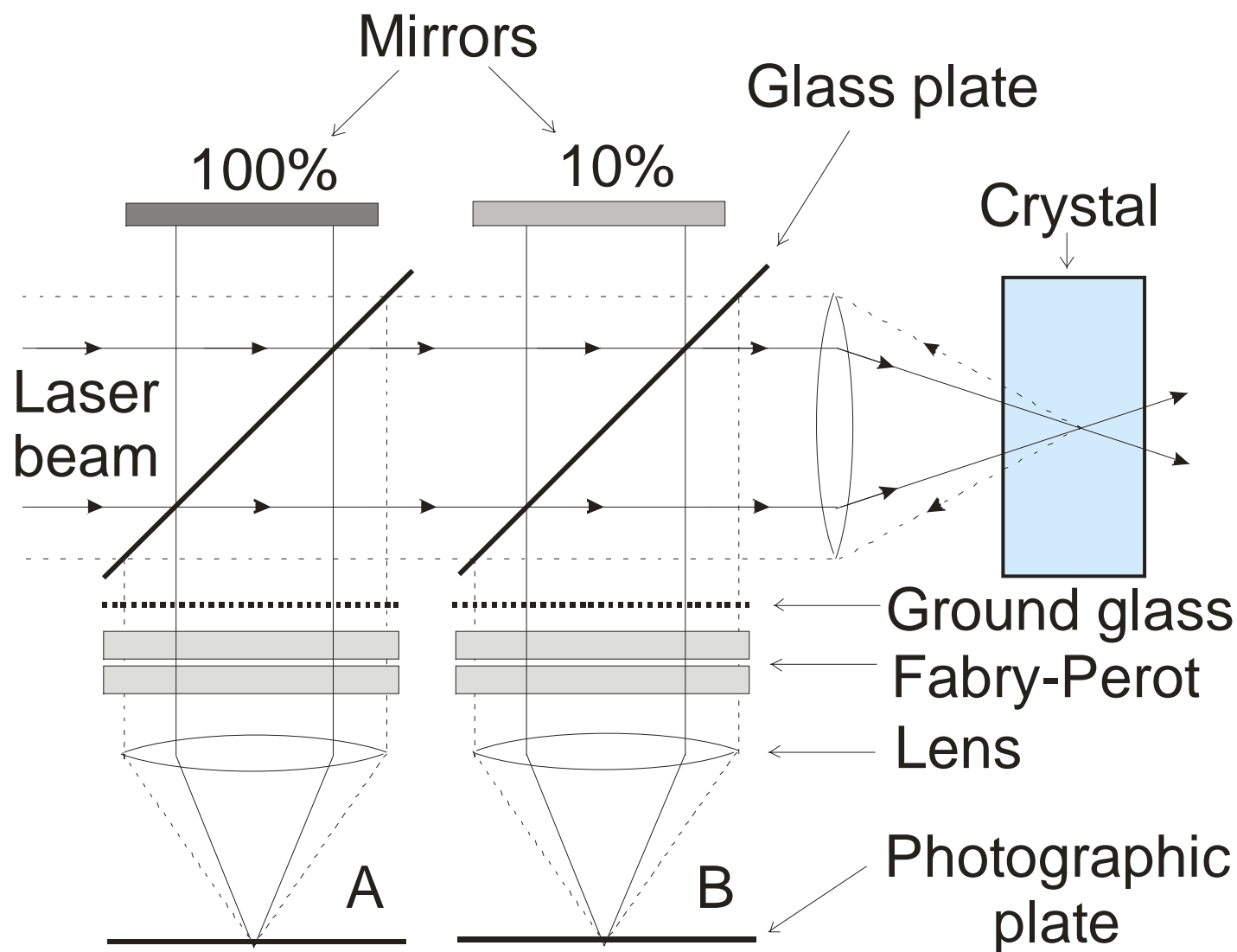
# Electrostriction couples sound waves to light waves



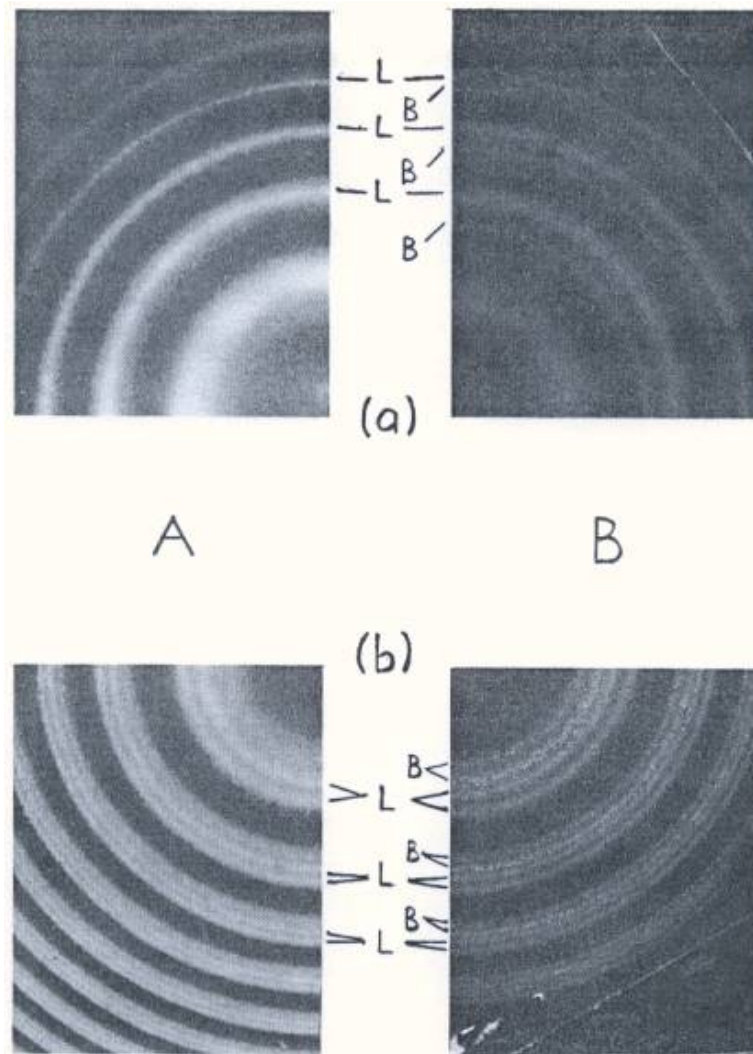
$$P_{\text{electrostriction}} = \left( \rho \frac{d\kappa}{d\rho} \right) \frac{1}{2} \varepsilon_0 E^2$$



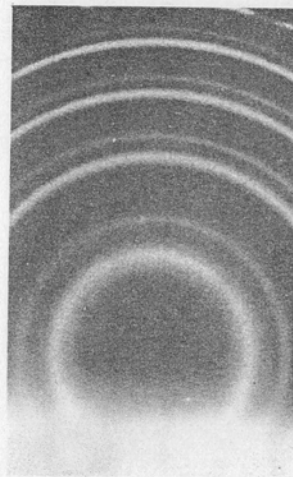
# Schematic of the Stimulated Brillouin Scattering Experiment



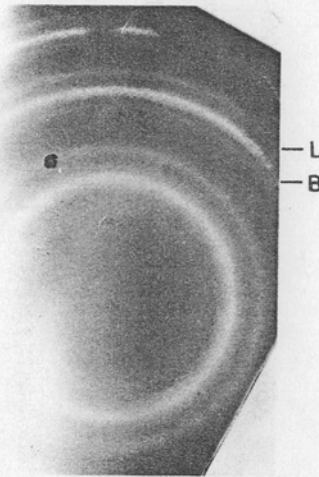
# Fabry-Perot Interferograms of SBS in Quartz



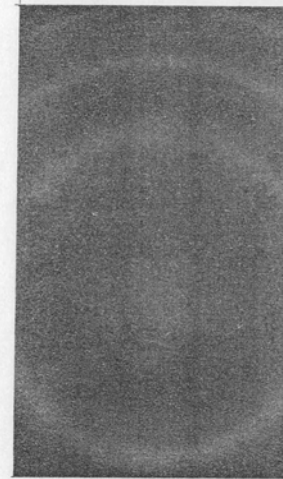
# SBS in Quartz and Sapphire



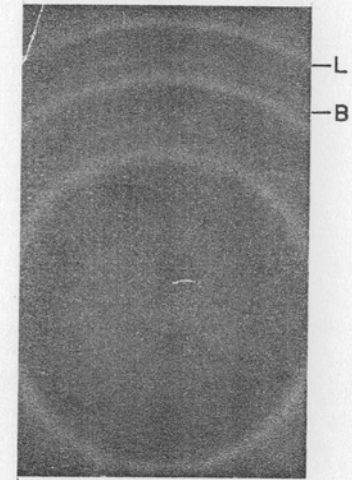
(a)



(b)



(c)



(d)

# Table of Observed and Predicted SBS Frequency Shifts in Quartz and Sapphire

photo part	crystal	scattering angle	laser incident along	SBS scattered along	laser polarized along	observed shift ( $\text{cm}^{-1}$ )	predicted shift ( $\text{cm}^{-1}$ )
(a)	Quartz	$180^\circ$	$Z$	$-Z$	$X$	0.99	0.97
(b)	Quartz	$180^\circ$	$X$	$-X$	$Z$	0.85	0.88
(c)	Quartz	$90^\circ$	$Z$	$\pm Y$	$X$	0.73	0.70
(d)	Sapphire	$180^\circ$	$Z$	$-Z$	$X$	2.07	2.01

# Applications of Stimulated Brillouin Scattering

- “Superluminal Propagation at Negative Group Velocity in Optical Fibers Based on Brillouin Lasing Oscillation”, by Liang Zhang (张亮), Li Zhan (詹黎), Kai Qian (钱楷), Jinmei Liu (刘金梅), Qishun Shen (沈启舜), Xiao Hu (胡晓), and Shouyu Luo (罗售余), Phys. Rev. Letters 107, 093903 (2011)

# Sommerfeld's velocities

- Phase velocity (can be superluminal)
- Group velocity (can also be superluminal)
- Front velocity (cannot be superluminal)

# Application to gravitational waves

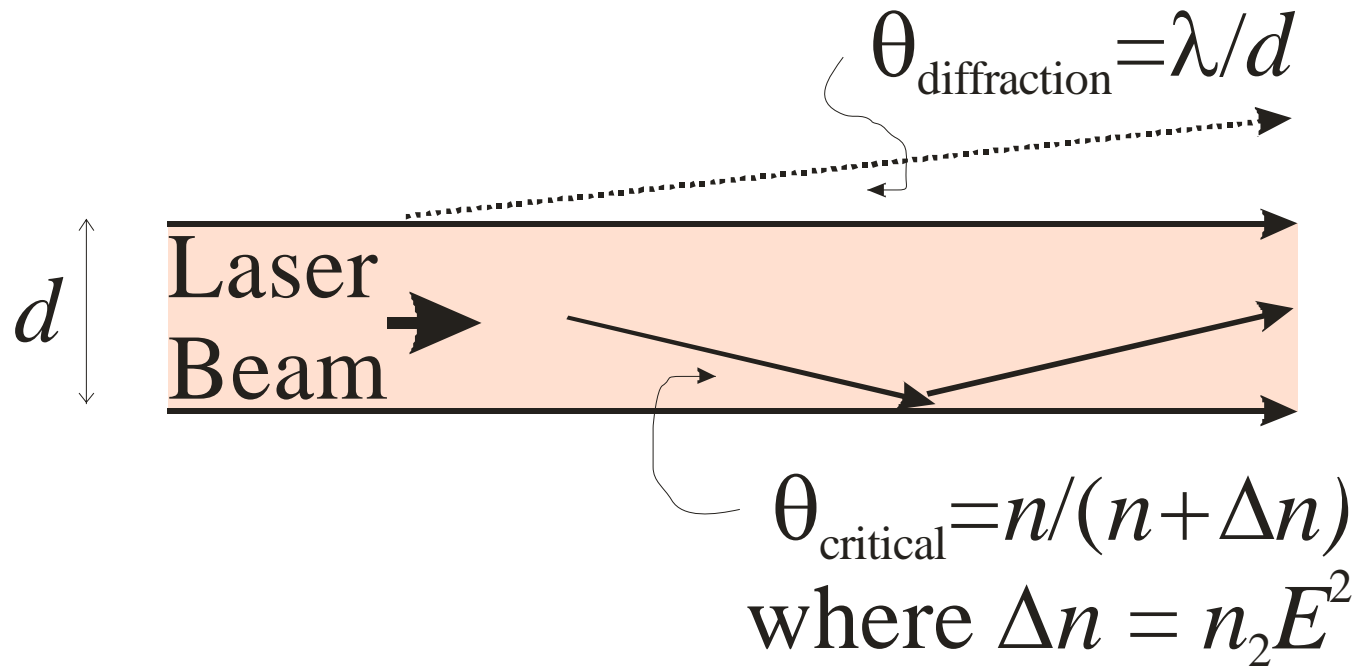
- *Superluminal* mass motions lead to the possibility of the *laboratory* generation of gravitational waves.
- Mirrors for gravitational waves are possible in the *linear* optics for these waves (Minter, Wegter-McNelly, and Chiao, Physica E 42, 234 (2010)).

# Self-trapping of optical beams

R. Y. Chiao, E. Garmire, and C. H.  
Townes, Phys. Rev. Letters 13,  
479 (1964).



# Townes's model for self-trapping



Self-trapping of laser beam

occurs when  $\theta_{\text{critical}} > \theta_{\text{diffraction}}$

# Ginzburg-Landau-like Equation

- For a slab-shaped laser beam, the wave equation in a nonlinear index medium becomes a Ginzburg-Landau-like equation

$$\frac{d^2 E}{dx^2} - \alpha E^2 + \beta E^3 = 0$$

is like

$$\frac{d^2 \Psi}{dx^2} - \alpha \Psi^2 + \beta \Psi^3 = 0$$

# Ginzburg-Landau-like Potential

$$(6.14) \quad V(x) = -\frac{1}{2} x^2 + \frac{1}{4} b x^4$$

which is drawn in fig. 28:

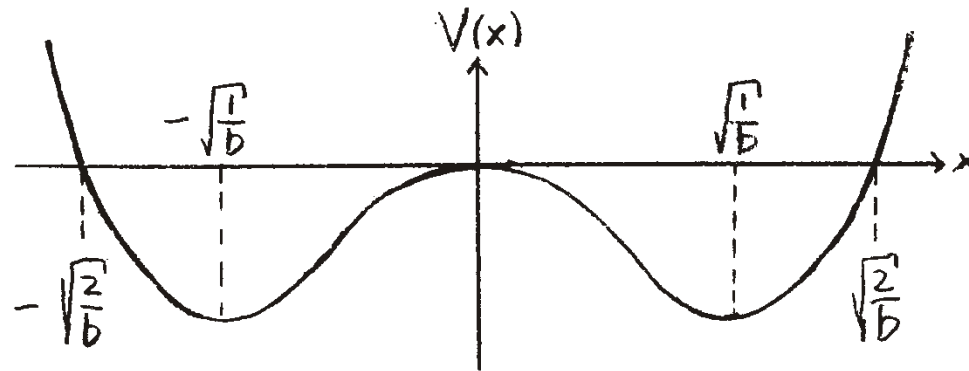


fig. 28: Potential for Mechanical Analogue to Self-Trapping

# Hyperbolic-Secant Spatial Soliton\*

$$E_{\text{soliton}}(x) = \left( \frac{2\alpha}{\beta} \right)^{1/2} \text{sech}(\alpha^{1/2}x)$$

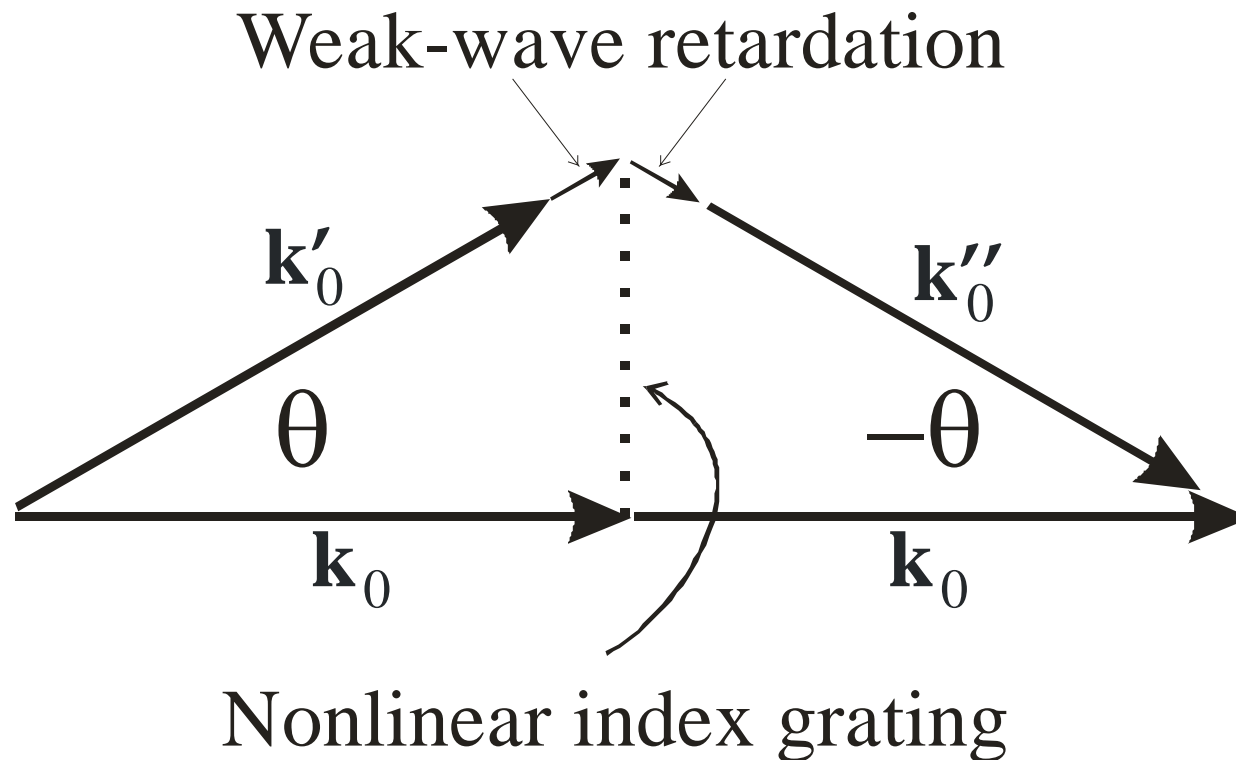
\* Chiao PhD thesis at MIT (1965)

- "Spatial Optical Solitons in Planar Glass Waveguides", Aitchison, Silberberg, Weiner, Oliver, Leaird, Jackel, Vogel, Smith, J. Opt. Soc. Am. B., 8, 1290 (1991).
- "Experimental Observation of Spatial Soliton Interactions", Aitchison, Weiner, Silberberg, Leaird, Oliver, Jackel, Smith, Opt. Lett., 16, 15 (1991).

# Degenerate Four-wave Parametric Amplification

- Bespalov & Talanov predicted degenerate four-wave parametric gain (Zh. Eksperim. i Teor. Fiz. – Pis'ma Redakt. 3, 307 (1966)).
- Chiao, Kelley, & Garmire showed that this gain originates from “weak-wave retardation” (PRL 17, 1158 (1966)).

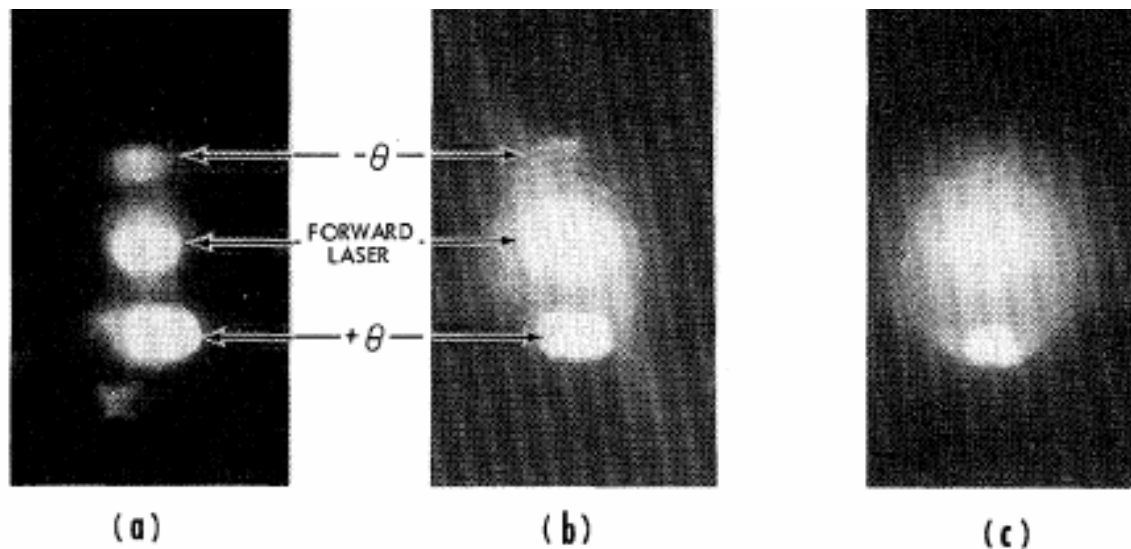
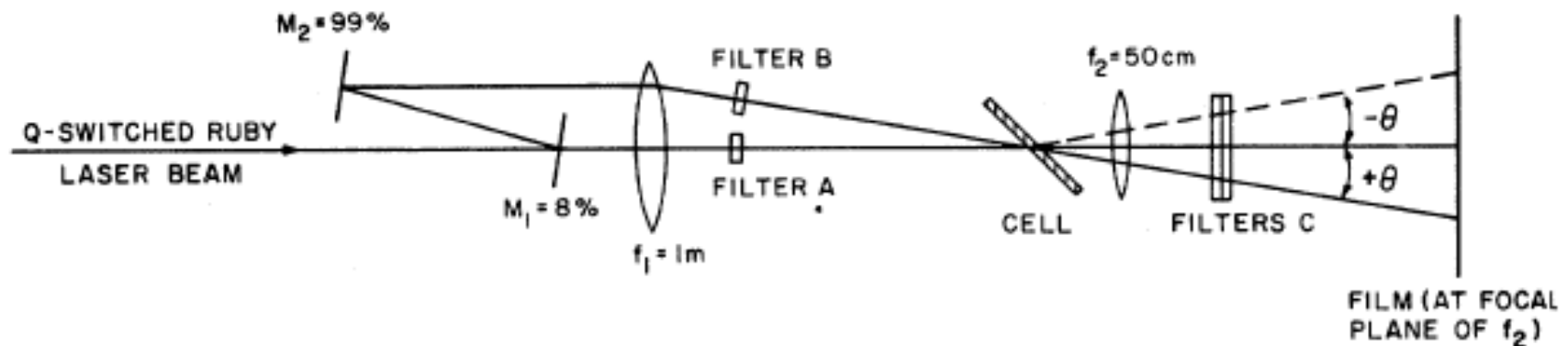
# Weak-wave retardation



Note that all four waves have the same frequency.  
Weak-wave retardation leads to *degenerate*  
four-wave parametric gain

# Observation of degenerate four-wave parametric amplification

- Carman, Chiao, & Kelley (PRL 17, 1281 (1966))



# Summary

- Stimulated Raman scattering in calcite obeying 3D phase-matching conditions was found in 1964.
- Stimulated Brillouin scattering was discovered in quartz and sapphire in 1964.
- The hyperbolic-secant 1D spatial soliton was theoretically predicted in 1964, and was observed in planar glass waveguides in 1991.
- Degenerate four-wave gain was seen in 1966.