INSTITUTE FOR RESEARCH IN ELECTRONICS & APPLIED PHYSICS

PHYS 761, Oct.22 (2019)

Nonlinear Optics

Raman Scattering Processes

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Blg. JMP , room 2202 , 11:00-12:15

Filling in for Prof. T. Antonsen

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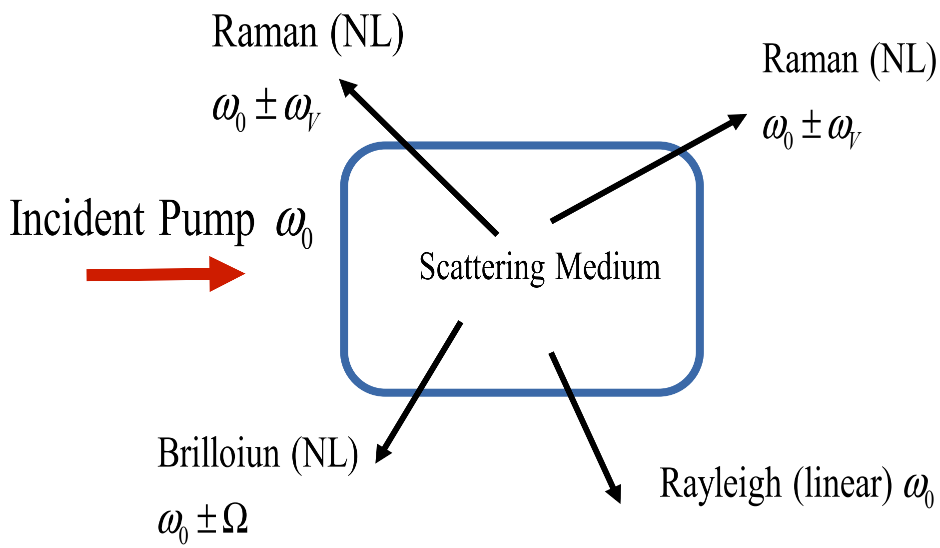
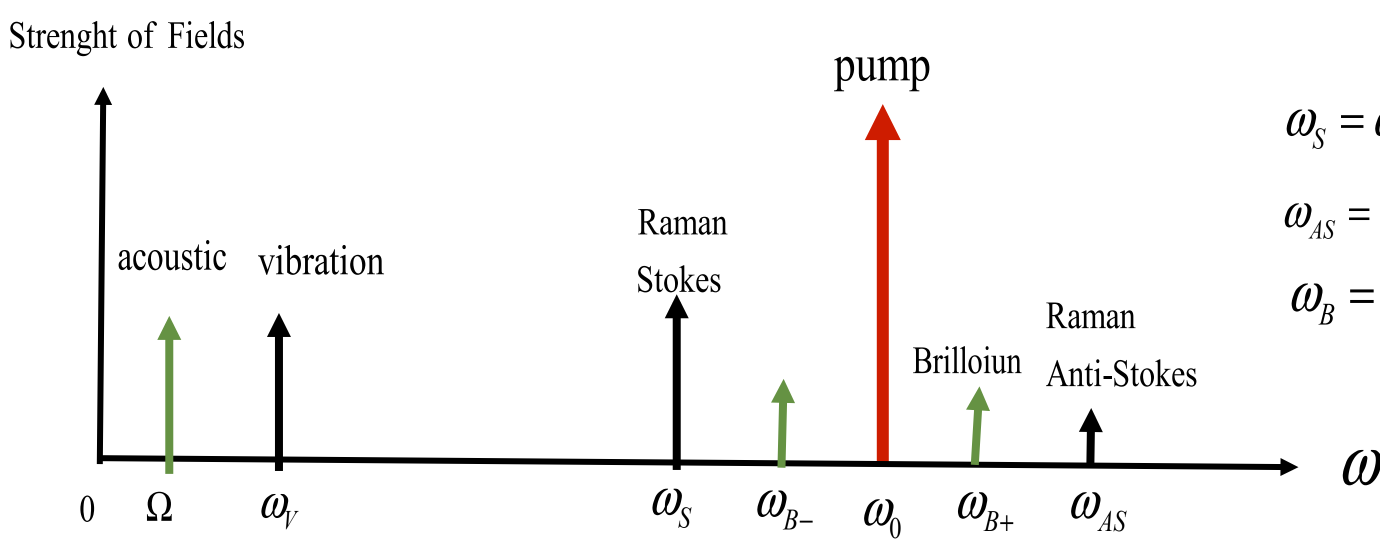
Outline

- Scattering Overview
- Raman Scattering
- Raman Plasma Instabilities
- Coherent Anti-Stokes Raman Scattering (CARS)

Nonlinear Scattering Processes





$$\omega_S = \omega_0 - \omega_P$$

$$\omega_{AS} = \omega_0 + \omega_P$$

$$\omega_B = \omega_0 \pm \Omega$$

Raman Scattering Processes

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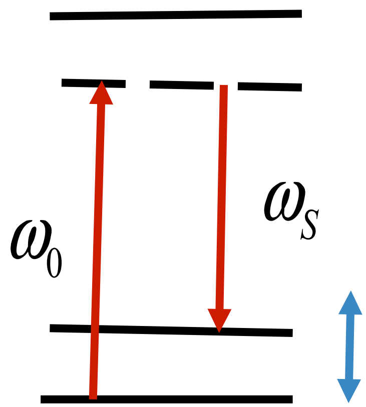
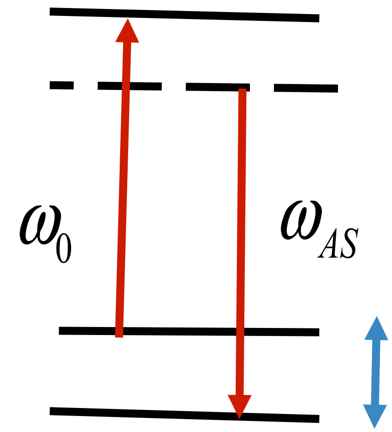
Energy Level Diagrams for Raman Stokes and Anti-Stokes Scattering

Stokes



$$\omega_V = \omega_0 - \omega_S$$

Anti-Stokes weak at low temperatures



$$\omega_V = \omega_{AS} - \omega_0$$

Raman Scattering: scattering off molecular **vibrational** modes, **rotational** modes, **plasma** waves or **ion accostic** waves, etc.

Brilllouin Scattering: scattering off induced density (sound/acoutic) waves by electrostriction

Classical Stimulated Raman Scattering Model

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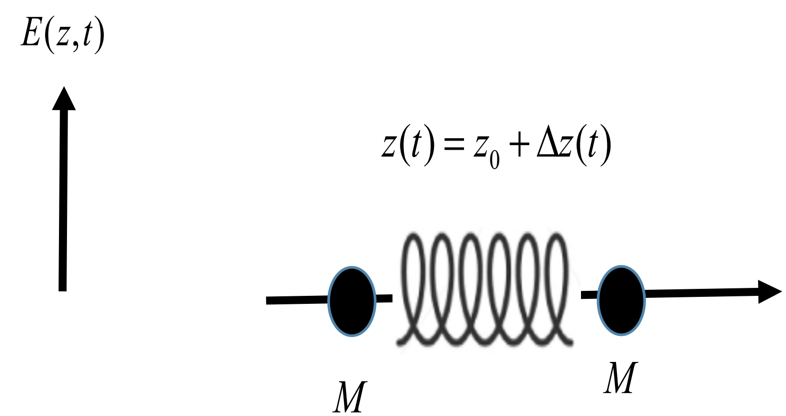
Assume that the optical field interacts with one of the vibrational modes of the molecules

$F(t)$ is a nonlinear force, e.g., ponderomotive force, which drives the vibrational mode of the molecule

$\Delta z(t)$ is the change in the intermolecular distance between the nuclear masses

The coherent vibational excitation of the molecules will coherently modulate the index of refraction of the medium

$$\frac{d^2\Delta z(t)}{dt^2} + 2\Gamma\frac{d\Delta z(t)}{dt} + \omega_V^2\Delta z(t) = \frac{F(t)}{M}$$



The material density is modulated at ω_V and therefore so is the refractive index

The nonlinear polarization field $P_{NL} = \varepsilon_0\chi_{NL}E$ contains many frequency components.

For example, $\omega_S = \omega_0 - \omega_V$ (Stokes) and $\omega_{AS} = \omega_0 + \omega_V$ (anti-Stokes) which drives the EM fields

Raman Instabilities in Plasmas

Raman processes and instabilities play important roles in many fields/areas of physics

- Laser fusion
- Laser driven particle acceleration
- Ioniospheric physics
- Raman amplifiers

Raman Instabilities in Plasmas

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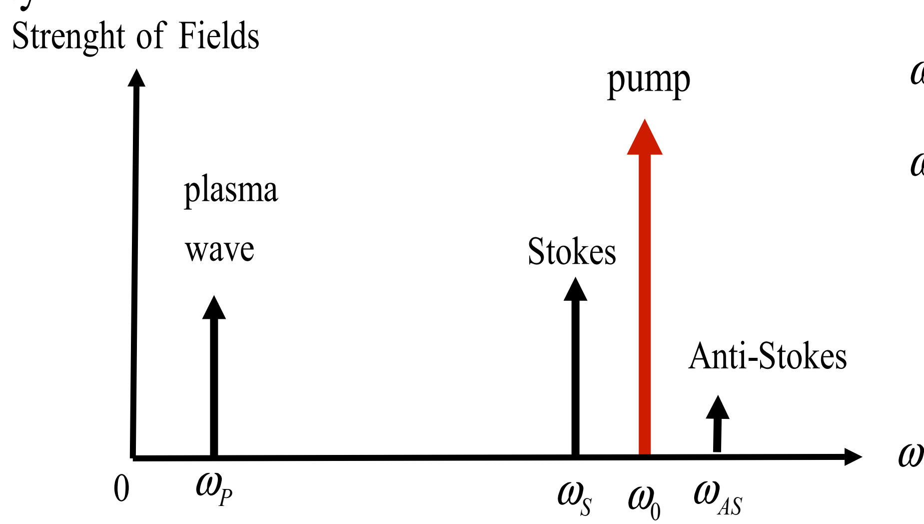
Consider the simplest case where the incident EM field (pump) is **mono-chromatic**, and nonrelativistic 1D fliud model.

In a laser-Raman interaction a large amplitude pump EM field beats with a low amplitude induced (noise) EM field.

The beating between these two EM fields resonantly excites a plasma (density) wave.

The plasma wave together with the transverse oscillation velocity due to the pump field provides a current source which excites (feeds, grows) the low amplitude EM field. This feedback leads to the Raman-Plasma instability



$$\omega_S = \omega_0 - \omega_P$$

$$\omega_{AS} = \omega_0 + \omega_P$$

Raman Dispersion Diagram

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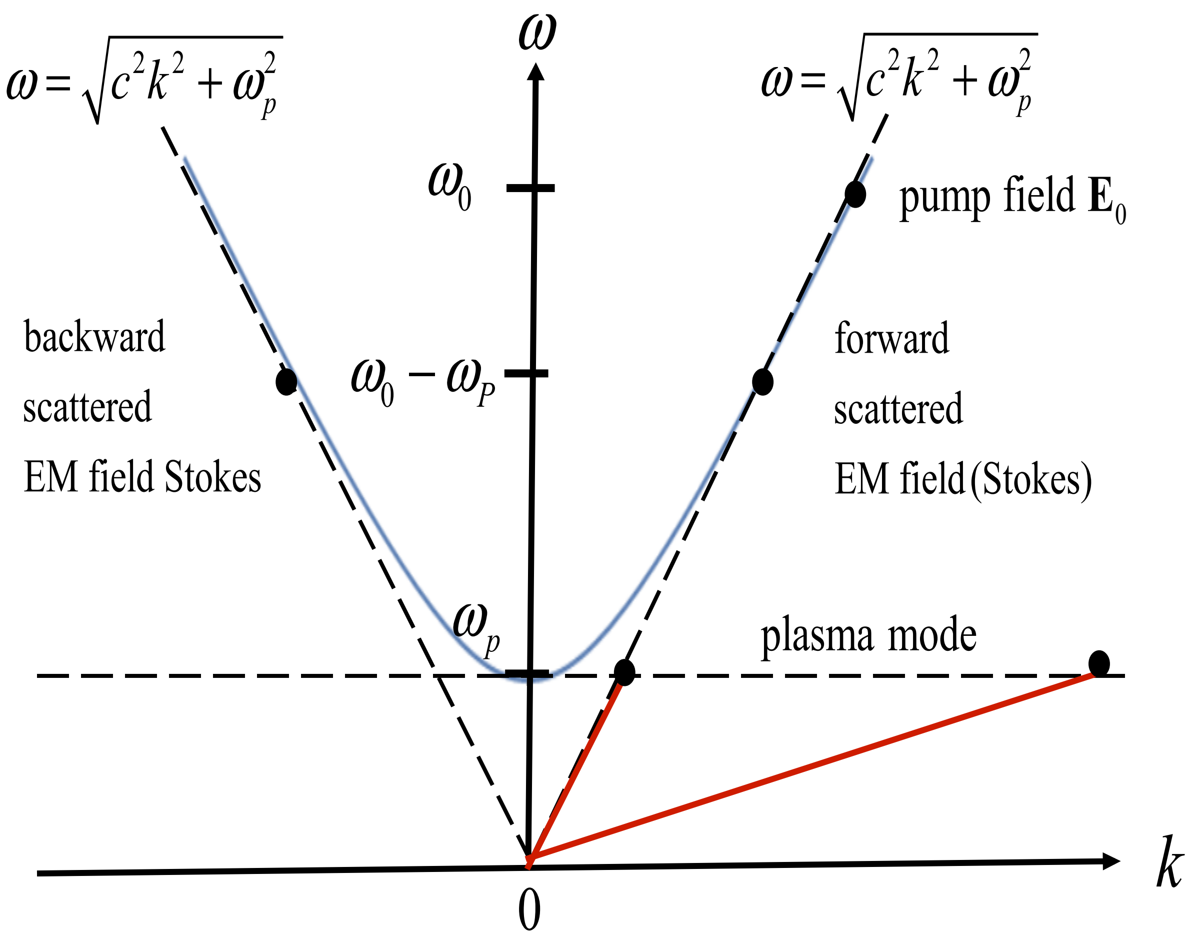


EM pump fields (circularly polarized):

$$\mathbf{E}_0(z,t) = \mathrm{Re}[E_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})] \;,$$

$$\mathbf{B}_0(z,t) = (k_0/\omega_0)\,\mathrm{Re}[-iE_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})]$$ where $k_0 = n_0\omega_0 / c$, ω_0 are real and E_0 is constant

It is convenient to use a circularly polarized rather than linearly polarized pump.

Poynting flux: $$\mathbf{S}_0 = \frac{1}{\mu_0}\mathbf{E}_0 \times \mathbf{B}_0 \;\rightarrow\; \mathbf{S}_0 = \frac{1}{\mu_0}\frac{k_0}{\omega_0}E_0^2(\cos^2(k_0 z - \omega_0 t) + \sin^2(k_0 z - \omega_0 t))\hat{\mathbf{z}} = c n_0 \varepsilon_0 E_0^2 \hat{\mathbf{z}}$$

note that $\mathbf{S}_0 = \langle \mathbf{S}_0 \rangle_{time}$

In a plasma: $$n_0 = \frac{ck_0}{\omega_0} = \sqrt{1 - \frac{\omega_p^2}{\omega_0^2}}$$

Raman Instabilities in Plasmas

Induced EM field (scattered):

$$\delta\mathbf{E}(z,t) = \mathrm{Re}[\delta E \exp(i(kz - \omega t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})] \ ,$$

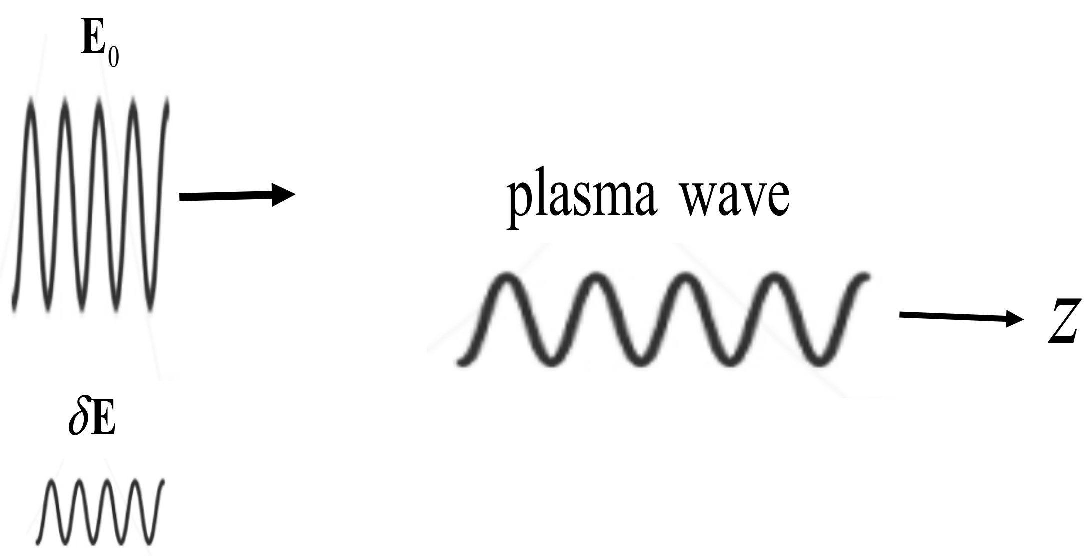
$$\delta\mathbf{B}(z,t) = \mathrm{Re}[-i(k/\omega)\delta E \exp(i(kz - \omega t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})]$$

where k, ω are complex and $|E_0| >> |\delta E|$

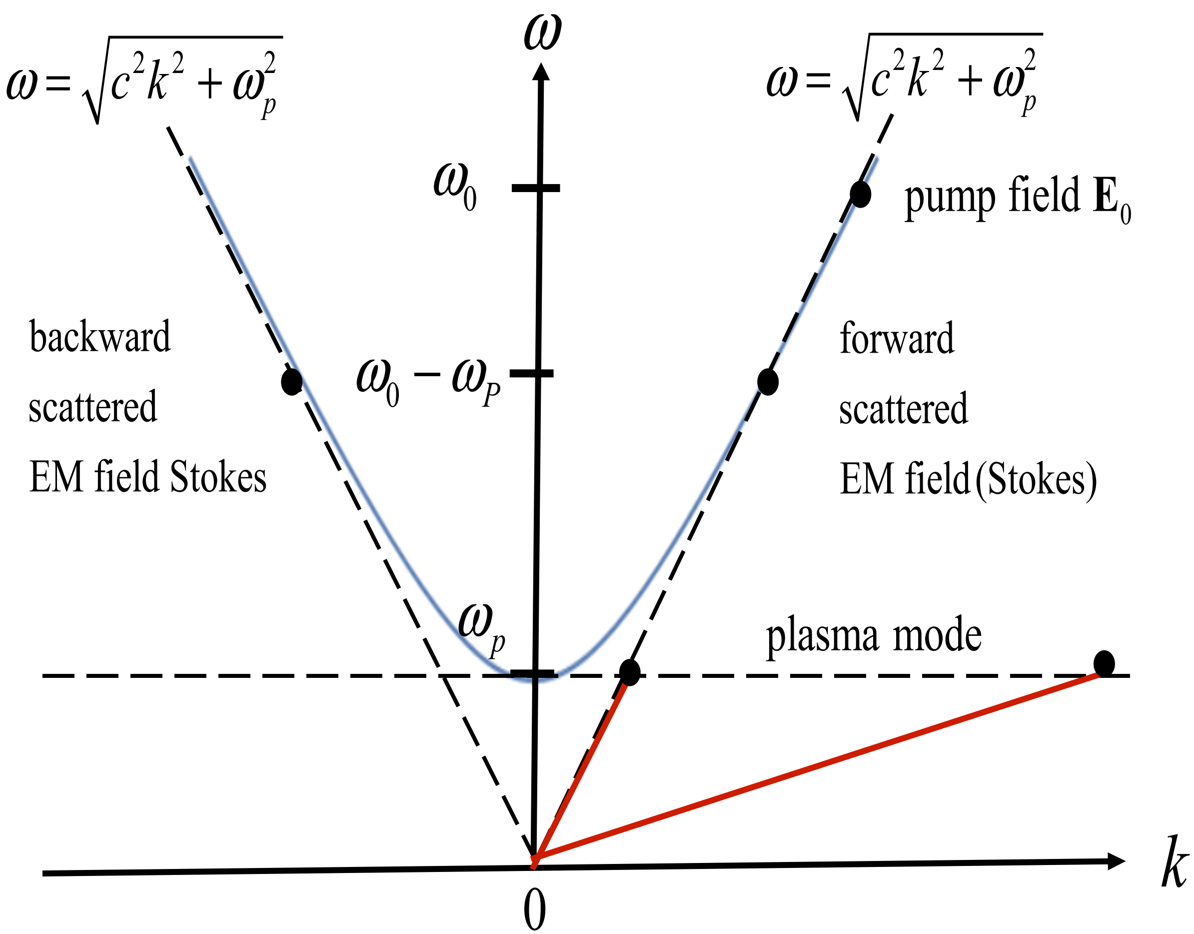
$\delta\mathbf{E}$ can be an input field or due to noise (spontaneous scattering)

$\delta\mathbf{E}$ can be forward or backward propagating

Induced Plasma wave (Stokes): $\delta\mathbf{E}_{plasma}(z,t) = \mathrm{Re}[\delta E_{plasma} \exp(i(k_0 - k)z - (\omega_0 - \omega)t)\hat{\mathbf{z}}]$



Raman Dispersion Diagram



Raman Dispersion Diagram

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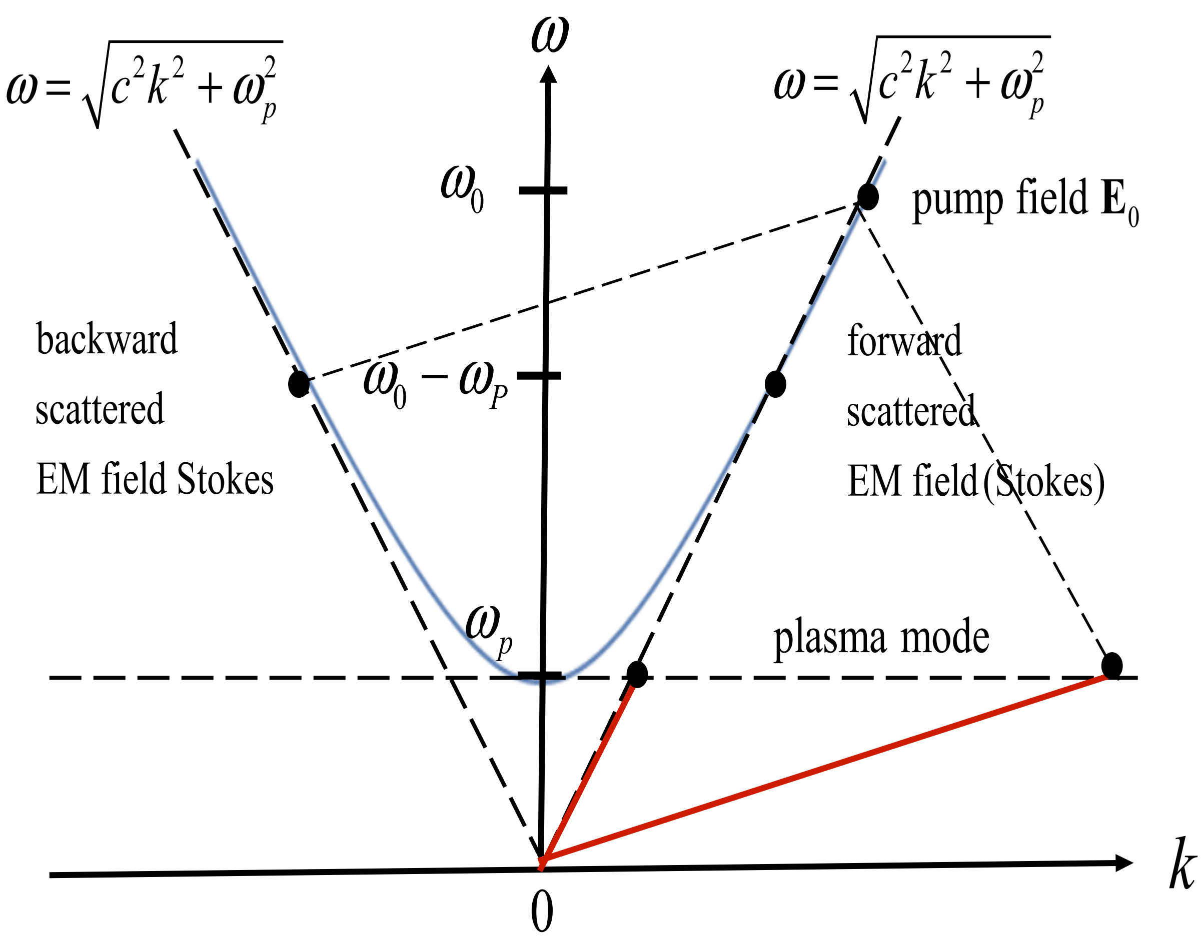

Raman Instabilities in Plasmas

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Goal: Determine the nonlinear **dispersion relation** $D(k,\omega)$ for the scattered field $\delta\mathbf{E}$

and growth rates for

Forward Raman: Stokes and Anti-Stokes scattering

Backward Raman: Stokes and Anti-Stokes scattering

Disperision relation (1D) of the **uncoupled** modes

Electromagnetic fields (pump and scattered): $\omega_0^2 - ck_0^2 - \omega_p^2 = 0$ and $\omega^2 - ck^2 - \omega_p^2 = 0$

Plasma wave: $(\omega_0 - \omega)^2 - \omega_p^2 = 0$

Dispersion relation of the **nonlinearly coupled** modes

$$\underbrace{\left(\omega^2 - ck^2 - \omega_p^2\right)}_{\text{EM mode}}\underbrace{\left((\omega_0 - \omega)^2 - \omega_p^2\right)}_{\text{Plasma mode}} = 2\omega_p^2\omega_0^2\beta_{osc}^2 \;\rightarrow\; \text{(nonlinear coupling)}$$

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The scattered field is given by $\left(\dfrac{\partial^2}{\partial z^2}-\dfrac{1}{c^2}\dfrac{\partial^2}{\partial t^2}\right)\delta\mathbf{E}(z,t)=\mu_0\dfrac{\partial\delta\mathbf{J}(z,t)}{\partial t}$

The induced current density $\delta\mathbf{J}$ is linear in $\delta\mathbf{E}$ and nonlinear in $\mathbf{E}_0$

The driving current $\delta\mathbf{J}$ is calculated to **1st order in the scattered field** δE
and contains terms up to **2nd order in the pump field** E_0

The polarization field **response** is assumed to be **instantaneous** and given by $\delta\mathbf{J}=\partial\delta\mathbf{P}/\partial t$

Polarization field: $\delta\mathbf{P}(\mathbf{r},t)=\varepsilon_0\left(\chi^{(1)}(\omega,k)+\chi^{(2)}(\omega_0,k_0,\omega,k)+\chi^{(3)}(\omega_0,k_0,\omega,k)\right)\delta\mathbf{E}(\mathbf{r},t)$

linear susceptibility
independent of $\mathbf{E}_0$

nonlinear
susceptibility $\sim(\mathbf{E}_0)^2$

Polarization field: $\delta\mathbf{P}(\mathbf{r},t)=\varepsilon_0\chi(\omega,k)\delta\mathbf{E}(\mathbf{r},t)$ where the refractive index is $n=\pm\sqrt{1+\chi}$

The induced response current density: $\delta\mathbf{J}(z,t)=qn(z,t)\mathbf{v}(z,t)\ \rightarrow$ terms of $\mathbf{Order}(\delta E)+\mathbf{Order}(E_0^2\delta E)$

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Fluid description of the plasma

Density eqn. : $$\frac{\partial n}{\partial t} + \frac{\partial (n \mathrm{v}_z)}{\partial z} = 0$$

Momentum eqns. (nonrelativistic): $$\left(\frac{\partial}{\partial t} + \mathrm{v}_z \frac{\partial}{\partial z} \right) \mathbf{v}(z,t) = \frac{q}{m} \left(\mathbf{E} + \mathbf{v} \times \mathbf{B} \right)$$

no damping or ionization

where $\mathbf{E} = \mathbf{E}_0 + \delta\mathbf{E} + \delta\mathbf{E}_{plasma}$, $\mathbf{B} = \mathbf{B}_0 + \delta\mathbf{B}$ and $\delta\mathbf{E}_{plasma}$ is the space charge field associated with the induced plasma wave

Raman Instabilities in Plasmas

Fluid description of plasma: $\frac{\partial n}{\partial t} + \frac{\partial (n \mathrm{v}_z)}{\partial z} = 0$ $\left(\frac{\partial}{\partial t} + \mathrm{v}_z \frac{\partial}{\partial z}\right)\mathbf{v}(z,t) = \frac{q}{m}\left(\mathbf{E}_{plasma} + \mathbf{E} + \mathbf{v}\times\mathbf{B}\right)$

The plasma fluid **velocity** and **density** are expanded to **second order** in the pump $\mathbf{E}_0$ and **first order** in the scattered (induced) field $\delta\mathbf{E}$ since we want the third order susceptibility $\chi^{(3)}(\omega_0, k_0, \omega, k)$ which gives the induced driving current $\delta\mathbf{J} = qn\mathbf{v}$ correct to second order in $\mathbf{E}_0$ and first order in $\delta\mathbf{E}$

$$\mathbf{v}(z,t) = \mathbf{v}_0 + \delta\mathbf{v} \quad \rightarrow \quad \mathbf{v}_0 = \mathbf{v}^{(0)} + \mathbf{v}^{(1)} + \mathbf{v}^{(2)} \qquad \mathbf{v}^{(0)} = 0$$

$$n(z,t) = n_0 + \delta n \quad \rightarrow \quad n_0 = n^{(0)} + n^{(1)} + n^{(2)} \qquad n^{(0)} = n_{P0}$$

$n^{(0)} = n_{P0}$: ambient plasma density (uniform and constant)

Deriving current density for scattered field: $\delta\mathbf{J} = qn_0\delta\mathbf{v} + q\delta n\mathbf{v}_0$

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Expansion of fluid density and velocity

$$\frac{\partial n_0}{\partial t}+\frac{\partial (n_0 \mathrm{v}_{0z})}{\partial z}=0 \qquad \frac{\partial \delta n}{\partial t}+\frac{\partial (n_0 \delta \mathrm{v}_z+\delta n \mathrm{v}_{0z})}{\partial z}=0$$

$$\frac{\partial \mathbf{v}_0}{\partial t}+\mathrm{v}_{0z}\frac{\partial \mathbf{v}_0}{\partial z}=\frac{q}{m}\left(\mathbf{E}_0+\mathbf{v}_0\times\mathbf{B}_0\right) \qquad \frac{\partial \delta\mathbf{v}}{\partial t}+\mathrm{v}_{0z}\frac{\partial \delta\mathbf{v}}{\partial z}+\delta\mathrm{v}_z\frac{\partial \mathbf{v}_0}{\partial z}=\frac{q}{m}\left(\delta\mathbf{E}_{plasma}+\delta\mathbf{E}+\underbrace{\mathbf{v}_0\times\delta\mathbf{B}+\delta\mathbf{v}\times\mathbf{B}_0}_{\text{ponderomotive force}}\right)$$

Since the pump is circularly polarized the axial velocity oscillation driven by the pump vanishes

$\mathbf{v}_0\times\mathbf{B}_0=0$ for a circularly polarized pump $\qquad \mathbf{v}_{0z}=0$

$$\mathbf{v}_{0\perp}(z,t)=\frac{q}{m\omega_0}\mathrm{Re}\left[iE_0\exp(i(k_0 z-\omega_0 t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})\right]$$

Raman Instabilities in Plasmas

$$\frac{\partial^2 \delta n}{\partial t^2} + n_0 \frac{q}{m}\frac{\partial}{\partial z}\left(\delta \mathbf{E}_{plasma} + \left(\mathbf{v}_0 \times \delta \mathbf{B} + \delta \mathbf{v} \times \mathbf{B}_0\right)_z\right) = 0$$

$$\frac{\partial \delta \mathbf{E}_{plasma}}{\partial z} = \frac{q}{\varepsilon_0}\delta n$$

Ponderomotive force ($\mathbf{v}\times\mathbf{B}$ term) drives the plasma wave

$$\frac{\partial^2 \delta n}{\partial t^2} + \omega_P^3 \delta n = -n_0 \frac{q}{m}\frac{\partial}{\partial z}\underbrace{\left(\mathbf{v}_0 \times \delta \mathbf{B} + \delta \mathbf{v} \times \mathbf{B}_0\right)_z}_{\text{ponderomotive force}}$$

$$\mathbf{v}_{0\perp} \times \delta \mathbf{B} = \frac{q}{2m\omega_0}\left(-i\frac{k}{\omega}E_0^* \delta E \exp(-i(k_0 - k)z + i(\omega_0 - \omega)t) + c.c\right)\hat{\mathbf{z}}$$

$$\delta \mathbf{v}_\perp \times \mathbf{B}_0 = \frac{qk_0}{2m\omega_0}\left(\frac{i}{\omega}E_0^* \delta E \exp(-i(k_0 - k)z + i(\omega_0 - \omega)t) + c.c\right)\hat{\mathbf{z}}$$

$$\left(\mathbf{v}_{0\perp} \times \delta \mathbf{B} + \delta \mathbf{v}_\perp \times \mathbf{B}_0\right)_z = \frac{q}{2m\omega_0}\left(i\frac{(k_0 - k)}{\omega}E_0^* \delta E \exp(-i(k_0 - k)z + i(\omega_0 - \omega)t) + c.c\right)\hat{\mathbf{z}}$$

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Ponderomotive force term induces the plasma wave

$$\frac{\partial^2 \delta n}{\partial t^2} + \omega_P^3 \delta n = -n_0 \frac{q}{m} \frac{\partial}{\partial z} \left(\mathbf{v}_0 \times \delta \mathbf{B} + \delta \mathbf{v} \times \mathbf{B}_0 \right)_z$$

ponderomotive force

$$\frac{\partial^2 \delta n}{\partial t^2} + \omega_p^2 \delta n = -\frac{q n_0}{m} \frac{q}{2 m \omega_0} \left[\frac{(k_0 - k)^2}{\omega} E_0^* \delta E \exp(-i(k_0 - k) z + i(\omega_0 - \omega) t) + c.c. \right]$$

$\sim \pm \omega_P$

$$q \delta n(z,t) = \varepsilon_0 \omega_p^2 \left[\frac{(k_0 - k)^2}{(\omega_0 - \omega)^2 - \omega_p^2} \left(\frac{q E_0^*}{2 m \omega_0} \right) \frac{\delta E}{\omega} \exp(-i(k_0 - k) z + i(\omega_0 - \omega) t) + c.c. \right]$$

Raman Instabilities in Plasmas
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Transvese component of $\delta\mathbf{v}$ is needed to second order in $\mathbf{E}_0$

$$\frac{\partial \delta\mathbf{v}_\perp}{\partial t} = \frac{q}{m}\delta\mathbf{E} + \frac{q}{m}\delta\mathbf{v}_z \times \mathbf{B}_0 - \delta \mathrm{v}_z \frac{\partial \mathbf{v}_{0\perp}}{\partial z} \qquad \mathbf{B}_0 = (k_0/\omega_0)\,\mathrm{Re}[-iE_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})]$$

$$\frac{\partial \delta n}{\partial t} + n_0 \frac{\partial \delta \mathrm{v}_z}{\partial z} = 0$$

$$q\delta n = \varepsilon_0 \omega_p^2 \left[\frac{(k_0 - k)^2}{(\omega_0 - \omega)^2 - \omega_p^2} \left(\frac{qE_0^*}{2m\omega_0} \right) \frac{\delta E}{\omega} \exp(-i(k_0 - k)z + i(\omega_0 - \omega)t) + c.c. \right]$$

$$qn_0 \delta \mathrm{v}_z = \varepsilon_0 \omega_p^2 \left[\frac{(k_0 - k)(\omega_0 - \omega)}{(\omega_0 - \omega)^2 - \omega_p^2} \left(\frac{qE_0^*}{2m\omega_0} \right) \frac{\delta E}{\omega} \exp(-i(k_0 - k)z + i(\omega_0 - \omega)t) + c.c. \right]$$

$$\mathbf{v}_{0\perp} = \frac{q}{m\omega_0}\mathrm{Re}\left[iE_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})\right] \qquad \frac{\partial \mathbf{v}_{0\perp}}{\partial z} = -\frac{qE_0}{2m\omega_0} k_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}}) + c.c.$$

$$qn_0 \delta \mathrm{v}_z \frac{\partial \mathbf{v}_{0\perp}}{\partial z} = \varepsilon_0 \omega_p^2 \left[\frac{(k_0 - k)(\omega_0 - \omega)}{(\omega_0 - \omega)^2 - \omega_p^2} \left(\frac{qE_0^*}{2m\omega_0} \right) \left(-\frac{qE_0}{2m\omega_0} \right) k_0 \frac{\delta E}{\omega} \exp(i(kz - \omega)t)(\hat{\mathbf{x}} + i\hat{\mathbf{y}}) + c.c. \right]$$

Raman Instabilities in Plasmas

(DETAILS)

Transvese component of $\delta\mathbf{v}$ is needed to second order in $\mathbf{E}_0$

$$\frac{\partial \delta \mathbf{v}_\perp}{\partial t} = \frac{q}{m}\delta\mathbf{E} + \frac{q}{m}\delta\mathbf{v}_z \times \mathbf{B}_0 - \delta \mathrm{v}_z \frac{\partial \mathbf{v}_{0\perp}}{\partial z} \qquad \mathbf{B}_0 = (k_0/\omega_0)\,\mathrm{Re}[-iE_0 \exp(i(k_0 z - \omega_0 t))(\hat{\mathbf{x}} + i\hat{\mathbf{y}})]$$

$$\delta \mathrm{v}_z \frac{\partial \mathbf{v}_{0\perp}}{\partial z} = \frac{\varepsilon_0 \omega_p^2}{q n_0}\left[\frac{(k_0 - k)(\omega_0 - \omega)}{(\omega_0 - \omega)^2 - \omega_p^2}\left(\frac{qE_0^*}{2m\omega_0}\right)\left(-\frac{qE_0}{2m\omega_0}\right)k_0 \frac{\delta E}{\omega}\exp(i(kz - \omega)t)(\hat{\mathbf{x}} + i\hat{\mathbf{y}}) + c.c.\right]$$

$$\frac{q}{m}\delta\mathbf{v}_z \times \mathbf{B}_0 = \frac{\varepsilon_0 \omega_p^2}{q n_0}\left[\frac{(k_0 - k)(\omega_0 - \omega)}{(\omega_0 - \omega)^2 - \omega_p^2}\left(\frac{qE_0^*}{2m\omega_0}\right)\left(\frac{qE_0}{2m\omega_0}\right)(-k_0)\frac{\delta E}{\omega}\exp(i(kz - \omega)t)(\hat{\mathbf{x}} + i\hat{\mathbf{y}}) + c.c.\right]$$

$$\boxed{\frac{q}{m}\delta\mathbf{v}_z \times \mathbf{B}_0 - \delta \mathrm{v}_z \frac{\partial \mathbf{v}_{0\perp}}{\partial z} = 0}$$

Raman Instabilities in Plasmas

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$$\delta\mathbf{J}_\perp = qn_0\delta\mathbf{v}_\perp + q\delta n\mathbf{v}_{0\perp}$$

$$\delta\mathbf{v}_\perp(z,t) = \frac{q}{m}\mathrm{Re}[\frac{i}{\omega}\delta E\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})]$$

$$q\delta n = \varepsilon_0\omega_p^2\left[\frac{(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\left(\frac{qE_0^*}{2m\omega_0}\right)\frac{\delta E}{\omega}\exp(-i(k_0-k)z+i(\omega_0-\omega)t)+c.c.\right]$$

$$\mathbf{v}_{0\perp}(z,t) = \frac{q}{m\omega_0}\mathrm{Re}\left[iE_0\exp(i(k_0z-\omega_0t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})\right]$$

$$\delta\mathbf{J}_\perp = \varepsilon_0\omega_p^2\frac{i}{2\omega}\delta E\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})$$

$$+\varepsilon_0\omega_p^2\frac{(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\left(\frac{qE_0^*}{2m\omega_0}\right)\left(\frac{qE_0}{2m\omega_0}\right)i\frac{\delta E}{\omega}\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})+c.c.$$

$$\delta\mathbf{J}_\perp = \frac{i}{2}\varepsilon_0\omega_p^2\left(1+\frac{2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\left(\frac{q|E_0|}{2m\omega_0}\right)^2\right)\frac{\delta E}{\omega}\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}})+c.c.$$

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$$\delta\mathbf{J}_\perp = q n_0 \delta\mathbf{v}_\perp + q\delta n \mathbf{v}_{0\perp}$$

$$\delta\mathbf{J}_\perp = \frac{i}{2}\varepsilon_0 \frac{\omega_p^2}{\omega}\left(1 + \frac{2c^2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\beta_{osc}^2\right)\delta E \exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}}) + c.c.$$

where $\beta_{osc} = \dfrac{q|E_0|}{2m\omega_0 c}$: oscillation velocity normalized to c

Polarization field: $\delta\mathbf{P}(\mathbf{r},t) = \varepsilon_0\left(\chi^{(1)}(\omega,k) + \chi^{(3)}(\omega_0,k_0,\omega,k)\right)\delta\mathbf{E}(\mathbf{r},t)$ $\quad\quad \delta\mathbf{J}_\perp = \partial\delta\mathbf{P}/\partial t$

$$\delta\mathbf{P} = \frac{\delta P}{2}\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}}) + c.c. = -\frac{\varepsilon_0}{2}\frac{\omega_p^2}{\omega^2}\left(1 + \frac{2c^2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\beta_{osc}^2\right)\exp(i(kz-\omega t))(\hat{\mathbf{x}}+i\hat{\mathbf{y}}) + c.c.$$

Linear and nonlinar susceptibility: $\chi^{(1)} = -\dfrac{\omega_p^2}{\omega^2}$ and $\chi^{(3)} = -2\dfrac{\omega_p^2}{\omega^2}\dfrac{c^2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\beta_{osc}^2$

Refractive index: $n = \pm\sqrt{1+\chi^{(1)}+\chi^{(3)}}$

Raman Instabilities in Plasmas (Raman Dispersion Relation)

Scattered field: $\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\delta\mathbf{E}(z,t) = \mu_0 \frac{\partial \delta\mathbf{J}(z,t)}{\partial t}$

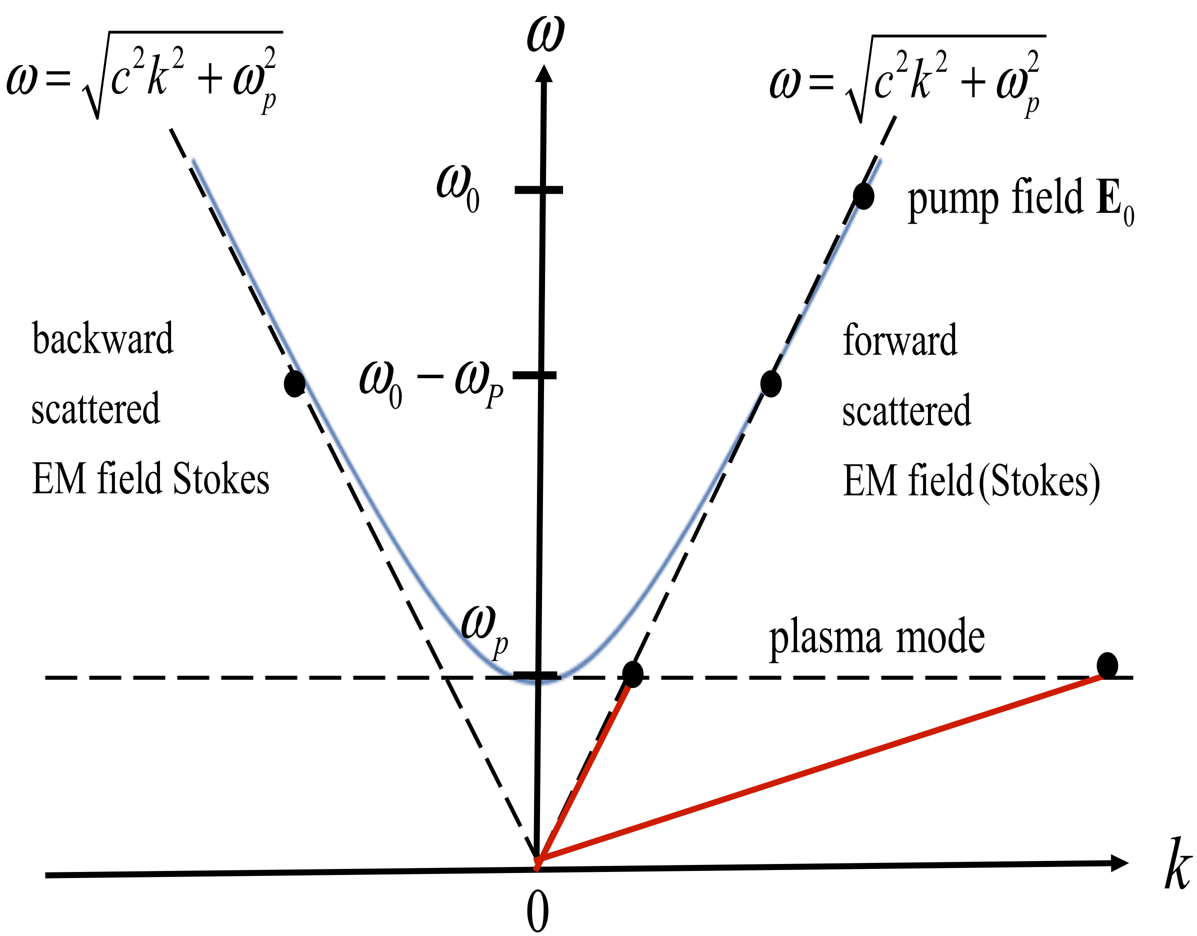
$$\left(-k^2 + \frac{\omega^2}{c^2}\right)\delta E = \mu_0\varepsilon_0\omega_p^2\left(1 + \frac{2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\left(\frac{q|E_0|}{2m\omega_0}\right)^2\right)\delta E \qquad \mu_0\varepsilon_0 = 1/c^2$$

$$\omega^2 - c^2k^2 - \omega_p^2 = 2\omega_p^2\beta_{osc}^2\frac{c^2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}$$

For the case where $\omega_0 >> \omega_P$

$$\underbrace{\left(\omega^2 - c^2k^2 - \omega_p^2\right)}_{\text{EM mode}}\underbrace{\left((\omega_0-\omega)^2 - \omega_p^2\right)}_{\text{Plasma mode}} = 2\omega_p^2\omega_0^2\beta_{osc}^2$$

Raman Dispersion Diagram



Raman Instabilities in Plasmas

Raman Dispersion Relation:

INSTITUTE FOR RESEARCH IN ELECTRONICS & APPLIED PHYSICS

$$(\omega_0-\omega)^2-\omega_p^2=\left((\omega_0-\omega)-\omega_P\right)\left((\omega_0-\omega)+\omega_P\right)\approx\begin{cases}2\omega_P\left((\omega_0-\omega)-\omega_P\right) & \text{Stokes}\\ -2\omega_P\left((\omega_0-\omega)+\omega_P\right) & \text{Anti-Stokes}\end{cases}$$

Consider the case where $\omega\approx\omega_0-\omega_P$ (Stokes) and $\omega_0>>\omega_P$

$$\omega^2-c^2k^2-\omega_p^2=2\omega_p^2\beta_{osc}^2\frac{c^2(k_0-k)^2}{(\omega_0-\omega)^2-\omega_p^2}\approx-\omega_P\beta_{osc}^2\frac{\omega_0^2}{\omega-(\omega_0-\omega_P)}$$

$$\left(\omega-\sqrt{c^2k^2+\omega_p^2}\right)\left(\omega-(\omega_0-\omega_P)\right)=-\frac{\omega_P\omega_0}{2}\beta_{osc}^2$$

$\omega=(\omega_0-\omega_P)+\Delta\omega$ $\qquad$ $\Delta\omega^2\approx-\frac{\omega_P\omega_0}{2}\beta_{osc}^2$ $\qquad$ $\Delta\omega=\pm i\sqrt{\frac{\omega_P\omega_0}{2}}\beta_{osc}$ **growing** and **decaying** wave

The Anti-Stokes scattering, $\omega\approx\omega_0+\omega_P$, is **stable**

Coherent Anti-Stokes Raman Spectroscopy (CARS)

Institute for Research in Electronics & Applied Physics
University of Maryland

CARS: Employs at two laser beams and relies on the third order nonlinearity $\chi^{(3)}$ of the material.

A **pump** field of frequency ω_0 and a **Stokes** field of frequency ω_S interact nonlinearly in a medium.

The two beams interact in the medium to produce a beat field at $\omega_V = \omega_0 - \omega_S$.

If the beat frequency is equal to one of the resonant **vibrational** or **rotational** frequencies in the medium the mode at ω_V is resonantly enhanced.

The resonantly enhanced vibrational mode beats with the Stokes field to generate an anti-Stokes field.

The anti-Stokes signal is the signature for the vibational mode.

Coherent Anti-Stokes Raman Spectroscopy (CARS)

CARS:

Employs at two laser beams and relies on the third order nonlinearity $\chi^{(3)}$ of the material.

Polarization field: $P = \varepsilon_0 \chi^{(1)} E + \varepsilon_0 \chi^{(3)} E^3$ $\qquad$ $J = \dfrac{\partial P}{\partial t} = \varepsilon_0 \chi^{(1)} \dfrac{\partial E(\mathbf{r},t)}{\partial t} + \varepsilon_0 \chi^{(3)} \dfrac{\partial E^3(\mathbf{r},t)}{\partial t}$

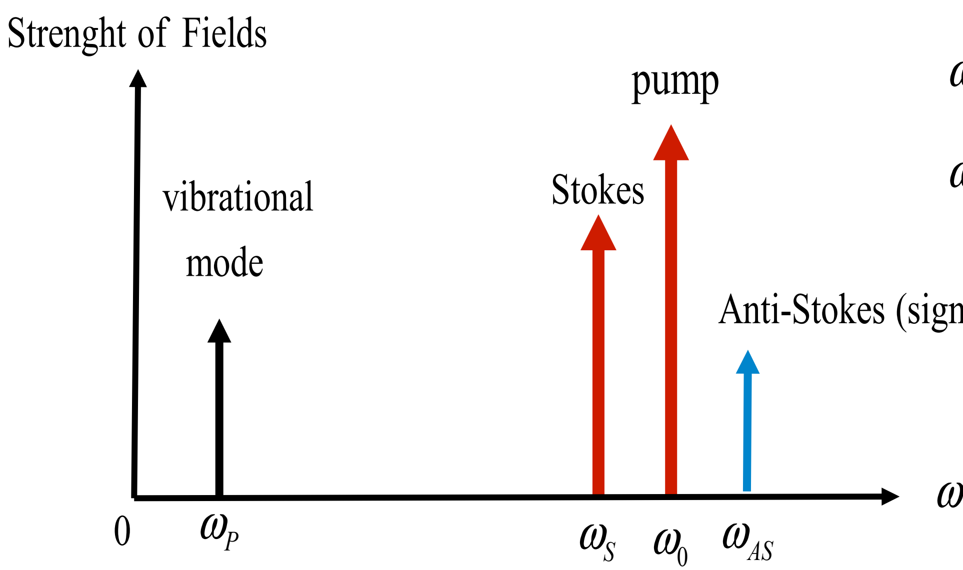
The susceptibilities $\chi^{(n)}$ are taken to be constant (instantaneous response)

The anti-Stokes field is given by $\left(\dfrac{\partial^2}{\partial z^2} - \dfrac{1}{c^2} \dfrac{\partial^2}{\partial t^2} \right) E_{AS} = \mu_0 \dfrac{\partial J_{AS}}{\partial t}$

Anti-Stokes field (signature): $\left(\dfrac{\partial^2}{\partial z^2} - \dfrac{(1+\chi^{(1)})}{c^2} \dfrac{\partial^2}{\partial t^2} \right) E_{AS} = \dfrac{\chi^{(3)}}{c^2} \dfrac{\partial^2}{\partial t^2} E_0 E_{probe} E_0$

Raman Instabilities in Plasmas

CARS



$$\omega_S = \omega_0 - \omega_P$$

$$\omega_{AS} = \omega_0 + \omega_P$$

END