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INTERNATIONAL CENTRE FOR THEORETICAL PHYSICS
I.C.T.P., P.O. BOX 586, 34100 TRIESTE, ITALY, CABLE CENTRATOM TRIESTE

SMR.769 - 13

WORKSHOP ON "NON-LINEAR ELECTROMAGNETIC INTERACTIONS IN SEMICONDUCTORS"

1 - 10 AUGUST 1994

"Semiconductor Bulk Media and Surfaces"

E. BURSTEIN
Physics Department
University of Pennsylvania
Philadelphia, PA 19104-6396
U.S.A.

These are preliminary lecture notes, intended only for distribution to participants

Inelastic Light Scattering by Electronic Excitations ("Electronic Raman Scattering")

A. Pinczuk
M.Y. Jiang
E. Burstein

Electronic Excitations

Intraband, Interband

Single Particle non-spin flip; collective Charge density

Single Particle spin-flip; collective spin density

Objectives

Elucidate physics underlying ILS as Phenomena

Use ILS as spectroscopic probe of electronic excitations $\omega(q)$ and via excitation profile measurements obtain information about electronic structure

① ILS by "free" carrier excitations in bulk (3-D) Semiconductors; Resonant ILS and mechanisms

② ILS by electronic excitations of 2-D confined electrons (holes) in surface space charge regions; hetero-structures (Quantum Wells and Superlattices).

Focus on <u>Resonant</u> Inelastic Light Scattering (R-ILS)

Resonant Inelastic Light Scattering

Outline

Historical Perspective

Progress during the last 25 years

materials synthesis, MBE etc

Instrumentation

Theory (condensed matter)

Major Emphases

Focus on underlying concepts!

Microscopic mechanisms, Selection rules

Incident and Scattered Radiation within media correspond to polaritons.

ILS (electronic Raman Scattering) is inelastic scattering of light by s.p. and collective electronic excitations and both intraband and interband electronic excitations in Bulk and 2-D heterostructures

- 2-photon process
- $\begin{cases} \text{2-step} \\ \text{3-step} \end{cases}$

Resonant ILS - greatly enhanced scattering intensities. Enables one to observe normally forbidden (e-R) scattering processes.

$$\langle (\phi u)_\beta | H_{int} | (\phi u)_\alpha \rangle \Rightarrow \overset{\text{interband}}{\langle u_\beta | H_{int} | u_\alpha \rangle} \langle \phi_\beta | \phi_\alpha \rangle + \underset{\text{intraband}}{\langle \phi_\beta | H_{int} | \phi_\alpha \rangle} \langle u_\beta | u_\alpha \rangle$$

polarizability

$$M(\omega_i) = \alpha_0 E(\omega_i)$$

oscillating electric dipole radiates $\longrightarrow$ Rayleigh Scattering $\boxed{\omega_s = \omega_i}$

(molecule) $$\alpha = \alpha_0 + \frac{\partial \alpha}{\partial u} u(\omega_v) + \cdots$$

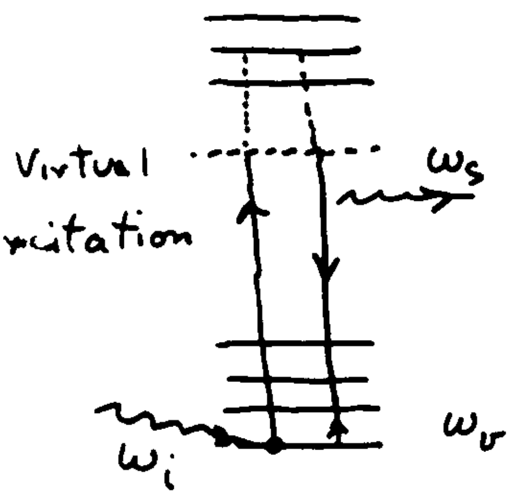
$$M(\omega_s) = \frac{\partial \alpha}{\partial u} u(\omega_v) E(\omega_i)$$ Raman Scattering

$$\boxed{\omega_s = \omega_i \mp \omega_v}$$

− Stokes
+ anti-Stokes

$$I(\omega_s) \propto \left| \frac{\partial \alpha}{\partial u} u \right|^2 I_0 \qquad I_0 \propto E_i^2$$

In terms of electronic and vibrational levels of molecule



- incident photon induces excitation of e to virtual intermediate state (induced elec. dipole). From virtual state e returns to ground electronic state but different vibrational level

$$\omega_i = \omega_s + \omega_v$$

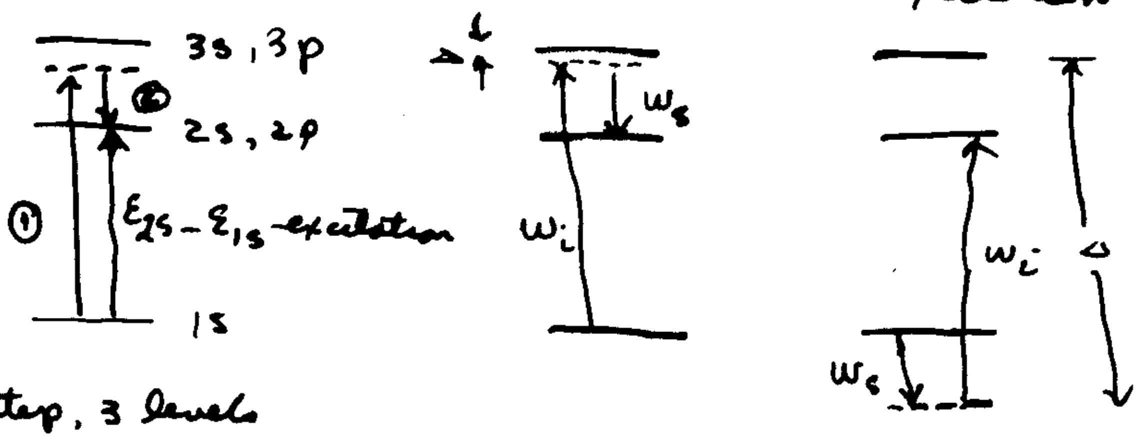
$$\omega_s = \omega_i - \omega_v \quad \text{Stokes RS}$$

Basics of Electronic Raman Scattering

Heitler "Quantum Theory of Radiation"

Raman scattering by atoms (H atom $1s \to 2s$)

excitation



2 step, 3 levels

2^{nd} order time dependent perturbation theory

$$\alpha_{1s\to 2s} \propto \frac{\langle 2s|p_s|3p\rangle\langle 3p|p_i|1s\rangle}{\mathcal{E}_{3p} - (\mathcal{E}_{1s} + \hbar\omega_i)} + \frac{\langle\,|\,|\,\rangle\langle\,|\,|\,\rangle}{\mathcal{E}_{3p} - (\mathcal{E}_{1s} - \hbar\omega_s)}$$

transition polarizability

THE POSSIBLE OBSERVATION OF ELECTRONIC RAMAN TRANSITIONS IN CRYSTALS

R. J. ELLIOTT
Clarendon Laboratory, Oxford, England

and

R. LOUDON
Royal Radar Establishment, Malvern, England

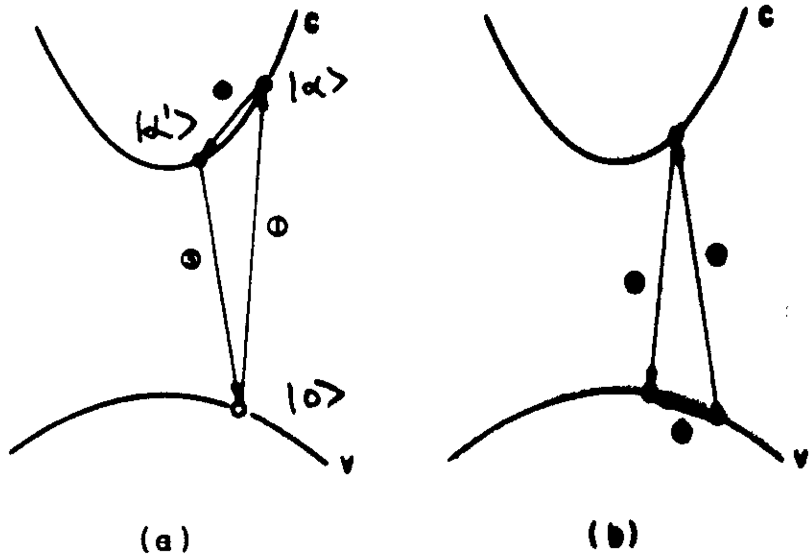
Received 8 December 1962

The Raman transition probability is obtained by second-order perturbation theory [4], and is proportional to the square of the matrix element for an isolated atom:

$$\langle \mathrm{i}, n_0, n_1 | F | \mathrm{f}, n_0-1, n_1+1 \rangle$$

$$= n_0^{\frac{1}{2}} \sum_j \left\{ \frac{\langle \mathrm{i} | \boldsymbol{\varepsilon}_0 \cdot \boldsymbol{p} | j \rangle \langle j | \boldsymbol{\varepsilon}_1 \cdot \boldsymbol{p} | \mathrm{f} \rangle}{E_j - E_\mathrm{i} - \hbar\omega_0} + \frac{\langle \mathrm{i} | \boldsymbol{\varepsilon}_1 \cdot \boldsymbol{p} | j \rangle \langle j | \boldsymbol{\varepsilon}_0 \cdot \boldsymbol{p} | \mathrm{f} \rangle}{E_j - E_\mathrm{i} + \hbar\omega_1} \right\} \quad (1)$$

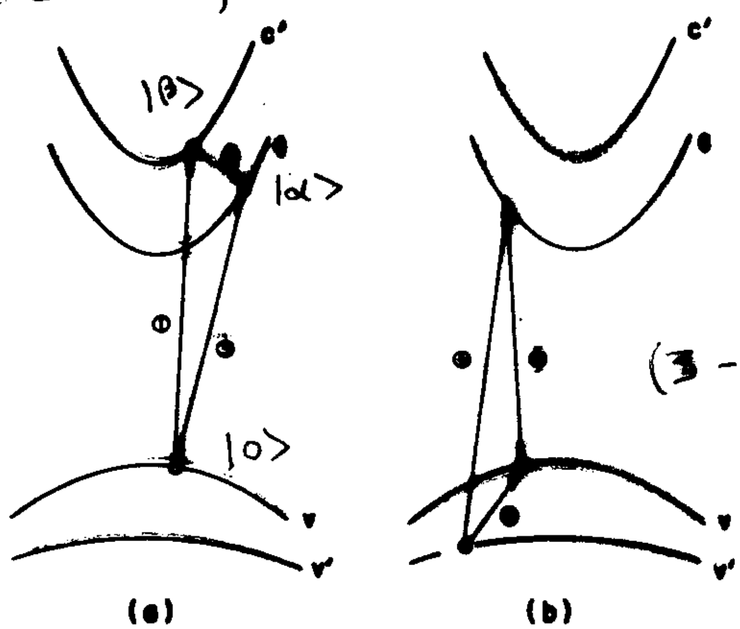
where n_0, n_1 are the occupation numbers of the incident and emitted photons with polarisation vectors $\boldsymbol{\varepsilon}_0$, $\boldsymbol{\varepsilon}_1$ and wave-vectors $\boldsymbol{q}_0$, $\boldsymbol{q}_1$. Finally $\boldsymbol{p}$ is the electronic momentum operator and j denotes the electronic intermediate states (excluding the initial and final states i and f).



(2-BAND RS)

Fig. 2. Schematic diagram of the interband and intraband transitions which play a role in *two band* scattering processes. The numbers 1 , 2 , and 3 indicate the time order of the electronic transitions.

R. LOUDON, Advances in Physics (1964)



3-Wave Mixing

vertical transitions

(3-BAND RS)

Fig. 3. Schematic diagram of the interband transitions which play a role in three band scattering processes. The numbers 1 , 2 , and 3 indicate the time order of the electronic transitions.

(H_{ep} = electron-phonon interaction)

$$R_j \propto \sum \frac{(p\cdot A_s)(H_{ep})(p\cdot A_i)}{(\mathcal{E}_{0\beta}-\hbar\omega_s)(\mathcal{E}_{\alpha 0}-\hbar\omega_i)} + 5 \text{ other terms}$$

$$R_j \propto \sum \frac{\langle 0|p\cdot A_s|\beta\rangle\langle\beta|H_{ep}|\alpha\rangle\langle\alpha|p\cdot A_i|0\rangle}{(\mathcal{E}_{0\beta}-\hbar\omega_s)(\mathcal{E}_{\alpha 0}-\hbar\omega_i)} \quad + \text{5 other terms}$$

$\alpha, \beta \equiv$ e-h pair states

$$\langle\beta|H_{ep}|\alpha\rangle = \Xi^{j}_{\beta\alpha} + F^{j}_{\beta\alpha} + A^{j}_{\beta\alpha}$$

$$(u_j) \qquad (E_j) \qquad (A_j)$$

<u>Frohlich</u>

$$F_{\alpha\alpha'}(k_j) = \pm i e \frac{E_j}{k_j}\,\delta(k'-k-k_j)\ ;\ \hat{e}_i \parallel \hat{e}_s$$

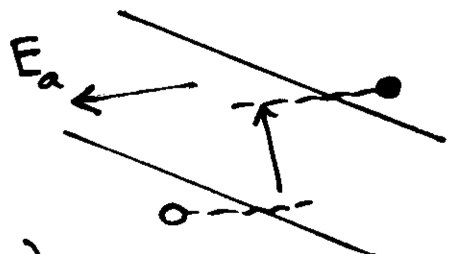
"diagonal" RS

intraband (2-BAND RS)

$$(F_{\alpha\alpha'})_e + (F_{\alpha\alpha'})_h = 0 \quad \text{for } k_j \to 0$$

(LOUDON)

<u>Homogeneous E_a</u>



$$(F_{\alpha\alpha'})_e + (F_{\alpha\alpha'})_h \propto r_{e-h}$$

$\vec{E}_j \parallel \vec{q}_j \parallel \vec{E}_a$ (ie, LO propagating $\parallel \vec{E}_a$) FRANZ-KELDYSH

<u>Inhomogeneous E_a</u> (also δ_{EM} very small)

large $k_j \Rightarrow \nabla E_j = i k_j E$

$$(F_{\alpha\alpha'})_e + (F_{\alpha\alpha'})_h \neq 0 \qquad \frac{\partial\chi}{\partial\nabla E_j}\nabla E_j = i k_j F E_j$$

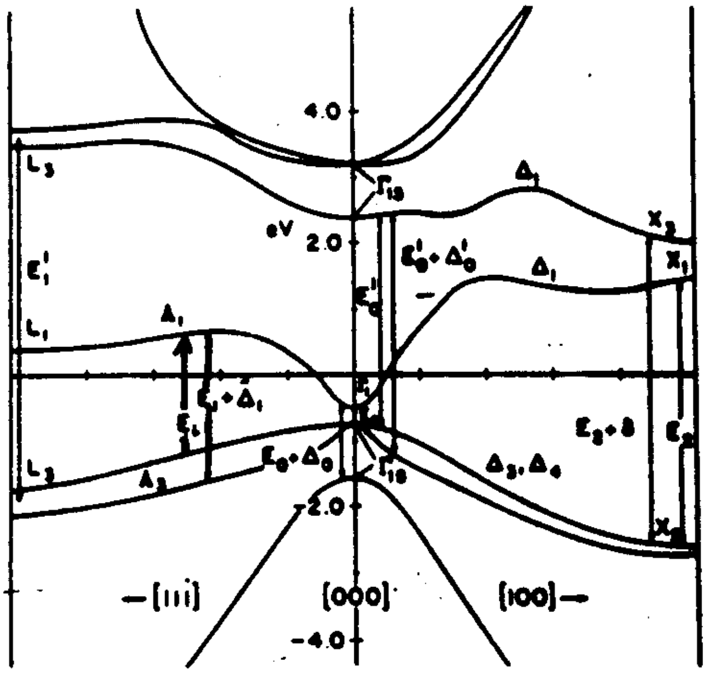


Fig. 7. Band structure of InSb showing the E_0 (Γ point), E_1 (along <111>) and E_2 (X point) critical points. [After F. H. Pollak, C. W. Higginbotham and M. Cardona, J. Phys. Soc. Japan, Suppl. 21, 20(1966).]

Raman Scattering at E_1 and $E_1+\Delta_1$ "gaps"

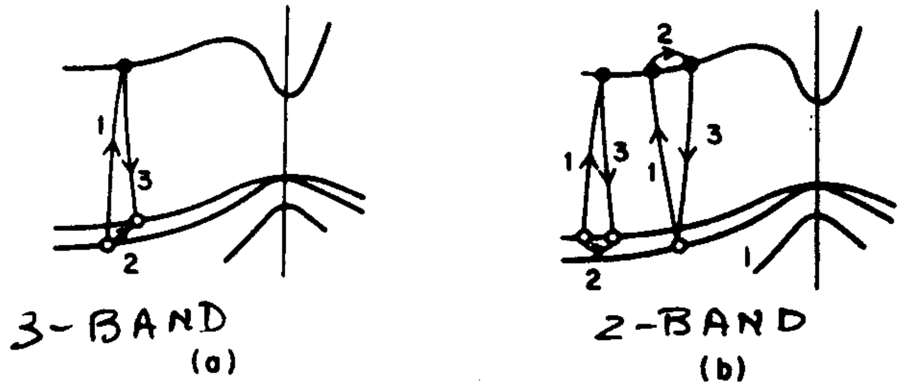
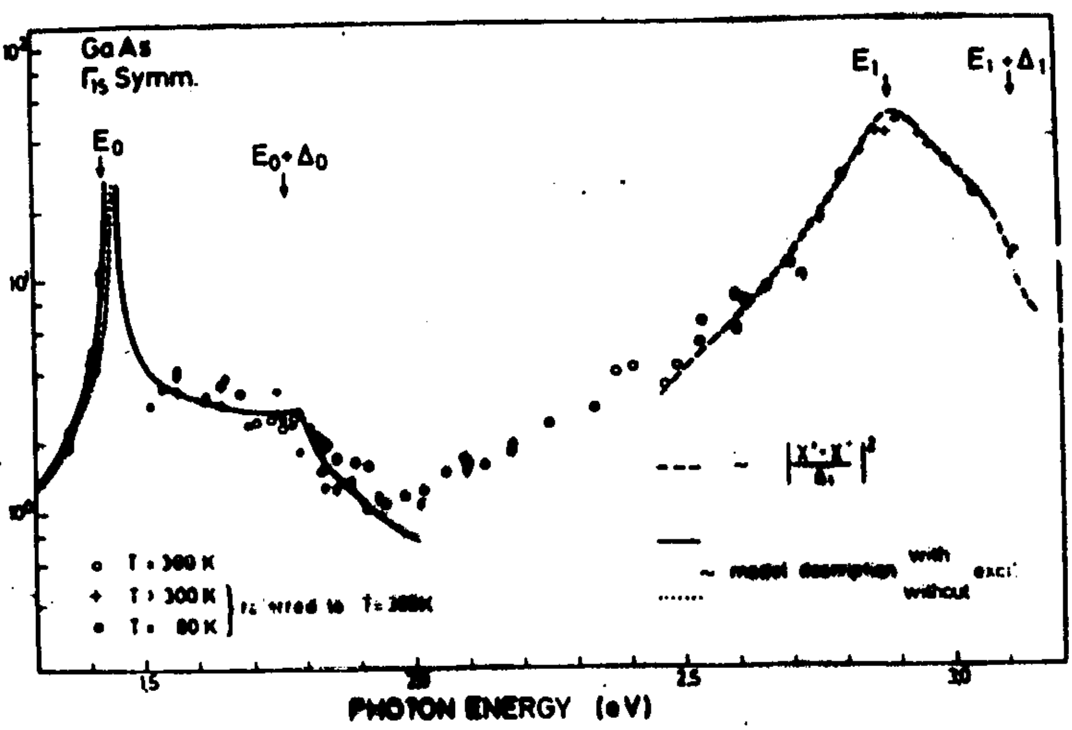


Fig. 6. Schematic diagrams showing (a) the transitions involved in the electro-optic scattering mechanism and in the 3-band deformation potential scattering mechanism, and (b) the transitions involved in the 2-band deformation potential and Fröhlich scattering mechanisms.

(Excitation Profile Measurement)



"RS is a form of Modulation Spectroscopy"
Cardona

RAMAN SCATTERING FROM InSb SURFACES AT PHOTON ENERGIES NEAR THE E_1 ENERGY GAP*

A. Pinczuk and E. Burstein
Physics Department and Laboratory for Research on the Structure of Matter,
University of Pennsylvania, Philadelphia, Pennsylvania
(Received 3 September 1968)

(110) Surface

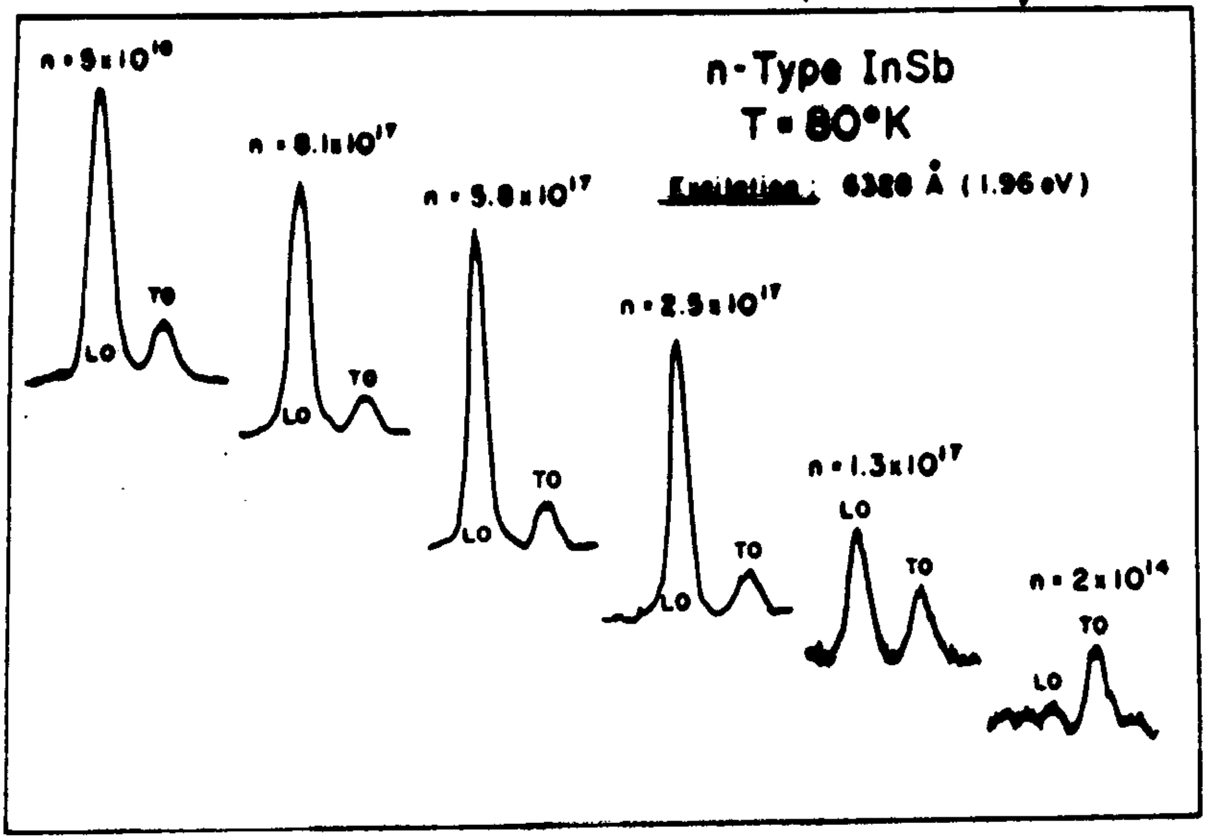
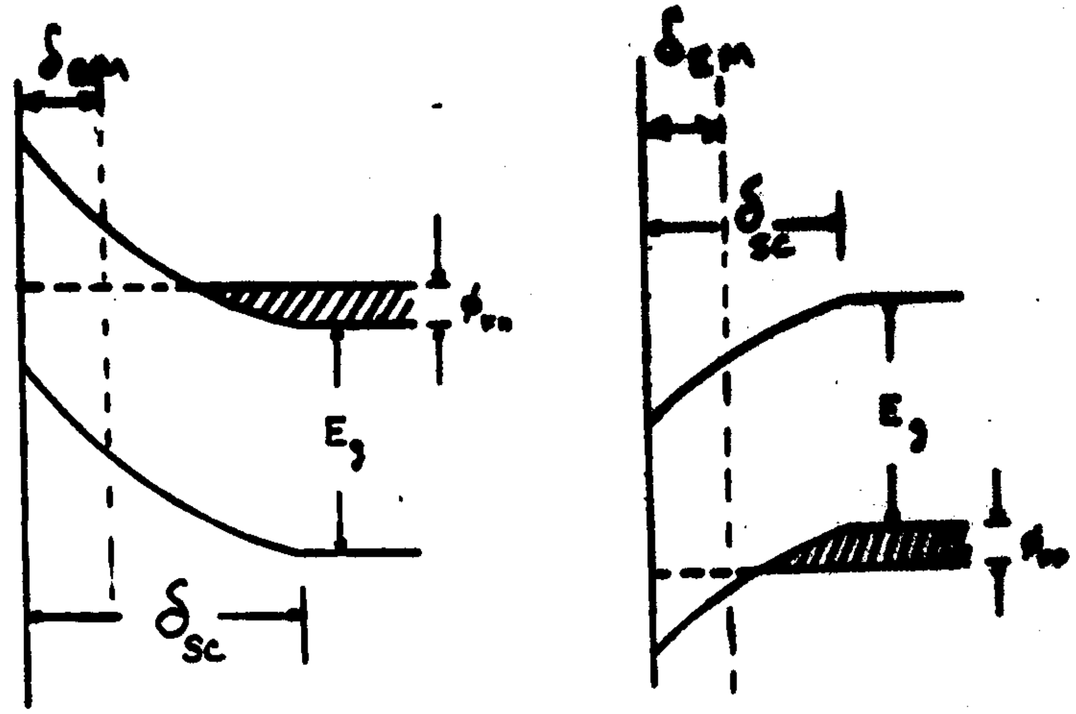


FIG. 2. Recorder traces of Raman spectra of LO and TO phonons of n-InSb at 80 °K.

LO forbidden

Surface Electric-Field—Induced Raman Scattering in PbTe and SnTe†

L. Brillson* and E. Burstein
Department of Physics and Laboratory for Research on the Structure of Matter,
University of Pennsylvania, Philadelphia, Pennsylvania 19104
(Received 20 May 1971)

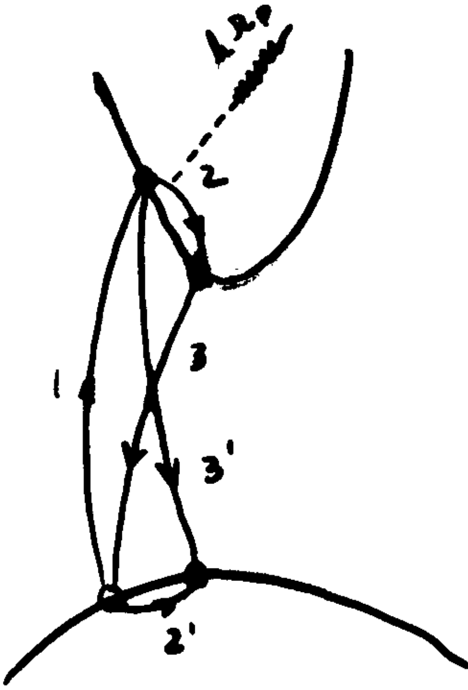


n-type
(a)

p-type
(b)

Raman Scattering Processes

$$\omega_i - \omega_s = \Omega$$
$$\vec{k}_i - \vec{k}_s = \vec{q}$$



"contact interaction"

Fröhlich (3 - [redacted])

e and h scattering contributions cancel in limit $q \to 0$

$$F_{\alpha\beta} = \pm i \langle\beta| e V e^{iq\cdot r}|\alpha\rangle \; ; \quad \hat{e}_i \parallel \hat{e}_s$$

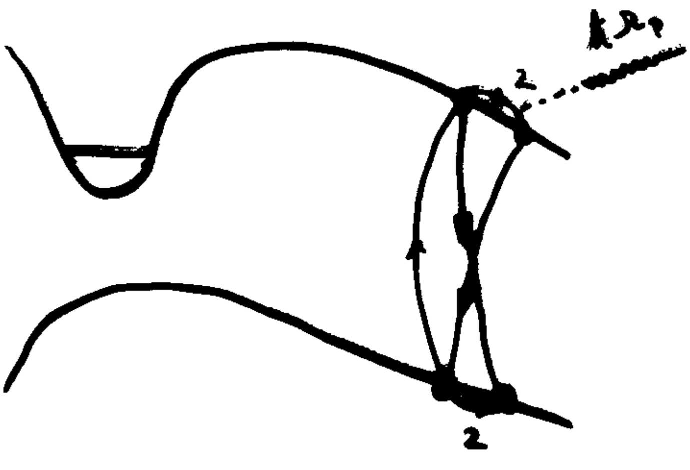
$I(q) \sim q^2$ providing $m_e \neq m_h$

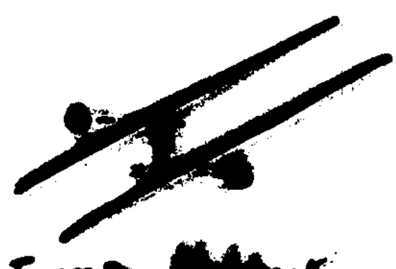
(wave vector dependent RS by elem. excitation)

$$I(E_A, q) \propto a q^2 + b E_A^2$$

$I(E_A) \neq 0$ in limit $q \to 0$

(Field induced RS by elem. excitation)
(Franz-[redacted])





Franz-[redacted]

A. RS by plasmons, LO phonons and by coupled LO phonon-plasmon modes $(\Omega_+, \Omega_-, \omega_L, \omega_p)$

$$\frac{d^2\sigma}{d\omega_s\, d\Theta} \propto \left| \sum_{\alpha} \frac{\langle 0|p_s|\beta\rangle\, F_{\alpha\beta} \langle\alpha|p_i|0\rangle}{(E_\alpha - \hbar\omega_i)(E_\beta - \hbar\omega_s)} \right|^2$$

2-Band (cont'd)

For excitons as intermediate state

$$\left(F_{x'x}\right) = \frac{ie\,E_{coul}}{K_{coul}}\left\{\langle x|e^{i(m_h/M)K_c\cdot R} - e^{i(m_e/M)K_c\cdot R}|x\rangle\right\}$$

For ~~hydrogenic~~ exciton $|x\rangle = |1s\rangle$

$$\left(F_{x'x}\right) = \frac{ie\,E_{coul}}{K_{coul}}\left(\frac{m_h - m_e}{M}\right)\left[1 - \left\{1 + \frac{1}{4}k_c^2 a_{1s}^2\right\}^{-2}\right]$$

For $m_e \neq m_h$ (and $K_c \neq 0$) $F_{x'x} \neq 0$

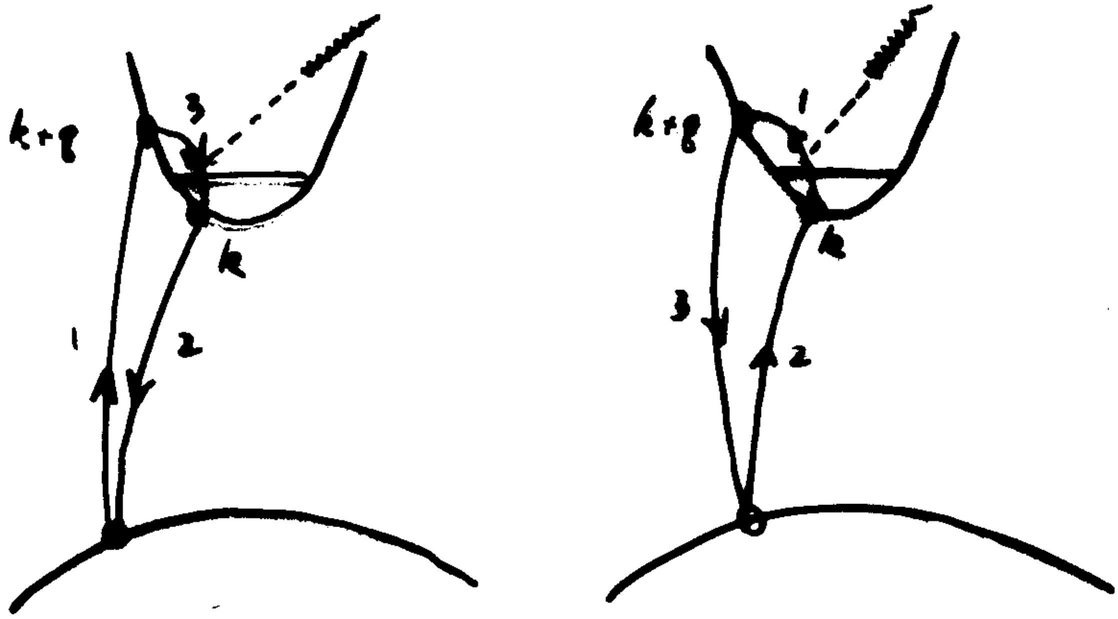
Note: $\left(A_{a'a}\right)_e = -\left(A_{a'a}\right)_h$ etc like F

However for $K \neq 0$, the ~~resultant~~ contribution is very small $\left(A_{a'a} = \frac{v}{c} F_{a'a}\right)$

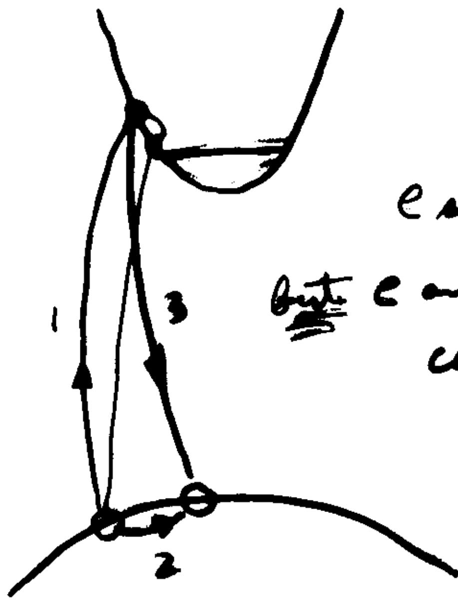
$\left(A_{x'x}\right) = 0$ even for $K \neq 0$ and $m_e \neq m_h$ for hydrogenic exciton

A. RS by collective excitations (Ω, q) (cont'd)

Hint: Σ, Γ and charge density



$$\frac{d^2\sigma}{d\omega_s\, d\Omega} \propto \left| \sum_{k'} \frac{\langle k|p|k'\rangle\, E_{k'k}\, \langle k'|p|0\rangle \; \left(f(k+q) - f(k)\right)}{\left(\mathcal{E}(k') - \hbar\omega_i\right)\left(e(k+q) - e(k) - \hbar\Omega\right)} \right|^2$$

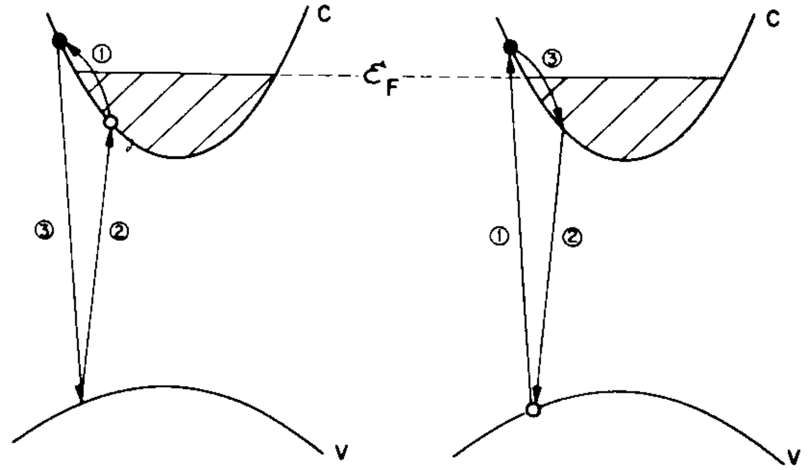


e scattering "blocked"

but e and h contributions do cancel in limit $q \to 0$

"Extreme Resonance" $\quad \hbar\omega_i = E_g + \frac{\hbar^2 k_F^2}{2m_v} + \frac{\hbar^2 k_F^2}{2m_c} = E_g'(n)$

Burstein and Pinczuk in "Physics of Opto-electronic Materials", edit by W. A. Albers Jr. (Plenum 19



(a) (b)

Fig. 6. Schematic diagram of interband and intraband transitions which play a role in two band scattering by plasmons in n-type semiconductors. The numbers 1 , 2 , and 3 indicate the time order of the electronic transitions.

$$R_j^{(1)}(\mathbf{q}) = V^{-1} \sum_{\mathbf{k}} \left\{ (p^i_{o\alpha})(p^s_{\beta o})(F^j_{\beta\alpha}) / [\omega_p + \mathcal{E}_c(\mathbf{k}+\mathbf{q}) - \mathcal{E}_c(\mathbf{k})] \right\} \times$$

$$\left\{ 2\omega_\alpha(\mathbf{k}) / [\omega_\alpha^2(\mathbf{k}) - \omega_1^2] \right\} [f(\mathbf{k}) - f(\mathbf{k}+\mathbf{q})],$$

$$(F^j_{\beta\alpha}) = e\, V(\mathbf{q})\, \delta(\mathbf{k}' - \mathbf{k} + \mathbf{q}) = i\, e\, [E_p(\mathbf{q})/q]\, \delta(\mathbf{k}' - \mathbf{k} + \mathbf{q})$$

$$\epsilon_T(q, \omega) = \epsilon(\omega) + \frac{4\pi e^2}{q^2} L_0$$

is the total dielectric constant including the electronic contribution. The scattering cross section is then

$$\frac{d^2\sigma}{d\omega d\Omega} = \left(\frac{e^2}{mc^2}\right)^2 \frac{1}{1-e^{-\omega/T}} \frac{1}{\pi}$$

$$\mathrm{Im} \left\{ \left[L_2 - \frac{4\pi e^2}{q^2} \frac{L_1^2}{\epsilon_T(q, \omega)} \right] (\hat{\epsilon}_1 \cdot \hat{\epsilon}_2)^2 + K_2 (\hat{\epsilon}_1 \times \hat{\epsilon}_2)^2 \right\}$$

Note that the spin-fluctuation contribution $K_2 (\hat{\epsilon}_1 \times \hat{\epsilon}_2)^2$ has no screening denominator.

Hamilton & McWhorter, in Light Scattering Spectra of Solids ed. by Wright & Mooradian (Springer 1969)

$$\frac{d^2\sigma}{d\Omega d\omega} \approx \left[\frac{E_g^2}{E_g^2 - (\hbar\omega_I)^2}\right]^2 \sigma_T^* (\vec{\epsilon}_I \cdot \vec{\epsilon}_F)^2 \frac{\hbar q^2 V}{4\pi^2 e^2} \epsilon_0^2(\omega) \operatorname{Im}\left[\frac{1}{\epsilon(\vec{q}, \omega)}\right]$$

$$\sigma_T^* = (e^2/m^* c^2)^2$$

$$\epsilon(0, \omega) \approx \epsilon_0(\omega) - \epsilon_\infty \frac{\omega_p^2}{\omega^2}$$

where

$$\epsilon_0(\omega) = \epsilon_\infty \left[\frac{\omega_{LO}^2 - \omega^2}{\omega_{TO}^2 - \omega^2}\right]$$

EXCITON-ENHANCED RAMAN SCATTERING BY OPTICAL PHONONS

E. Burstein*† and D. L. Mills‡
Physics Department, University of California, Irvine, California

and

A. Pinczuk* and S. Ushioda*
Physics Department and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania
(Received 16 September 1968)

The theory of exciton-enhanced Raman scattering is formulated in terms of the scattering of polaritons by optical phonons via the exciton part of the coupled modes. The expression for the exciton contribution to the scattering tensor is given, within a constant factor, in terms of the same parameters that determine the exciton contribution to the frequency-dependent dielectric constant. The theory also provides a new mechanism for the exciton contribution to the electro-optic effect.

Resonant Light Scattering by Single-Particle Electronic Excitations in *n*-GaAs†

A. Pinczuk,* L. Brillson, and E. Burstein
Department of Physics and Laboratory for Research on the Structure of Matter, University of Pennsylvania, Philadelphia, Pennsylvania 19104

and

E. Anastassakis
Department of Physics, Northeastern University, Boston, Massachusetts 02115
(Received 29 March 1971)

Resonant light scattering by single-particle excitations was observed in *n*-GaAs with incident photon energies near the $E_0 - \Delta_0$ optical energy gap. We find that under extreme resonance conditions the spectra have two components with the scattered light polarization perpendicular and parallel to the incident light. These results are interpreted in terms of a random-phase-approximation theory.

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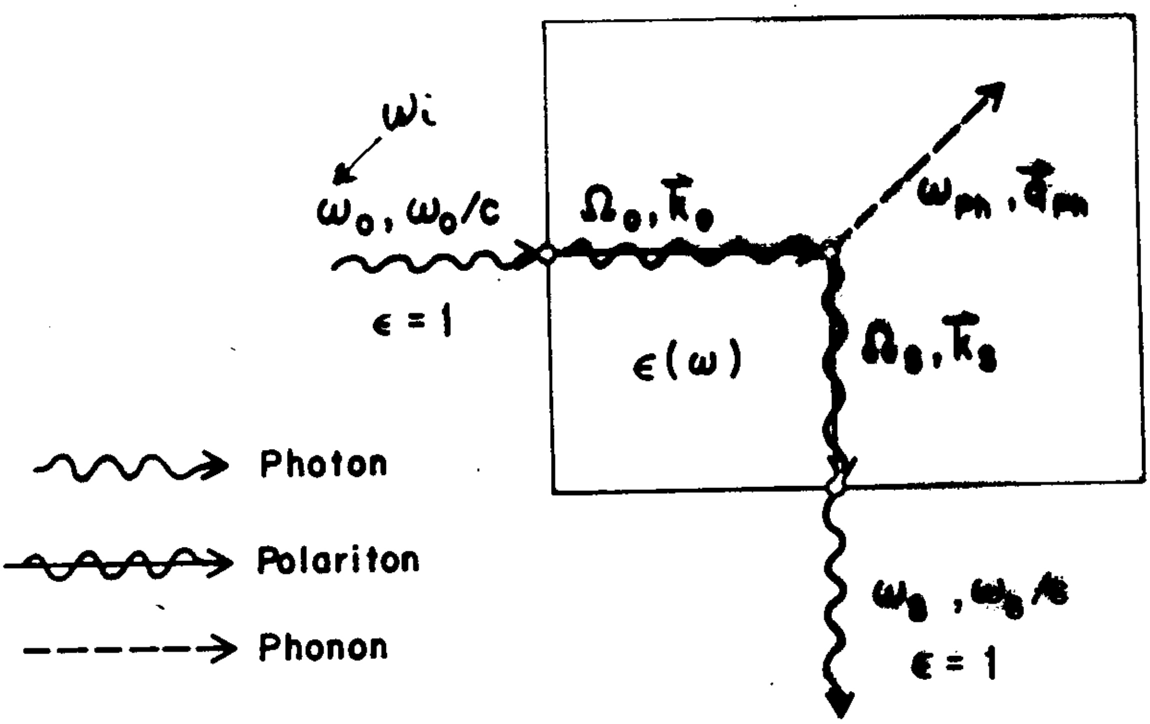
Conceptually very useful!

(Ovander)



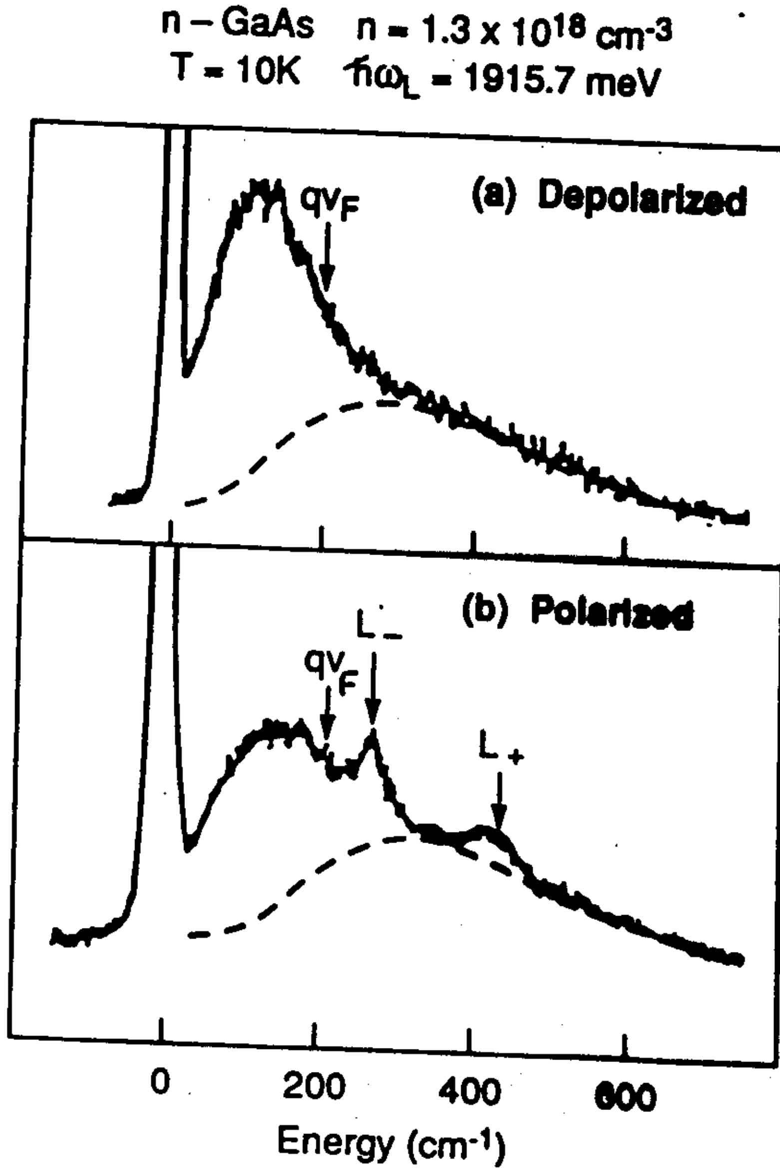
$$M_{nm} \sim \sum S_{xi}^{1/2} M_{is} S_{xs}^{1/2} \times F(\omega_{xi}, \omega_{xj}, \omega_i, \omega_s)$$

Polariton-phonon scattering matrix element

exciton-~~phonon~~ ~~matrix element~~
phonon induced transition between exciton levels)

$$M^{F}_{is} > M^{D}_{is}$$

Ovander
Mills et al
Bendow and Birman
~~Hopfield~~
Loudon

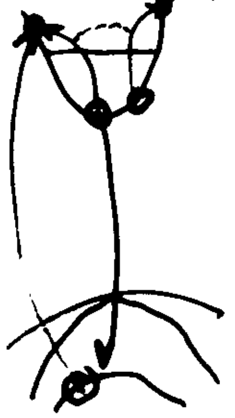

n – GaAs n = 1.3 x 10^18 cm^-3
T = 10K ħω_L = 1915.7 meV
qv_F
(a) Depolarized
(b) Polarized
L_–
qv_F
L_+
0
200
400
600
Energy (cm^-1)

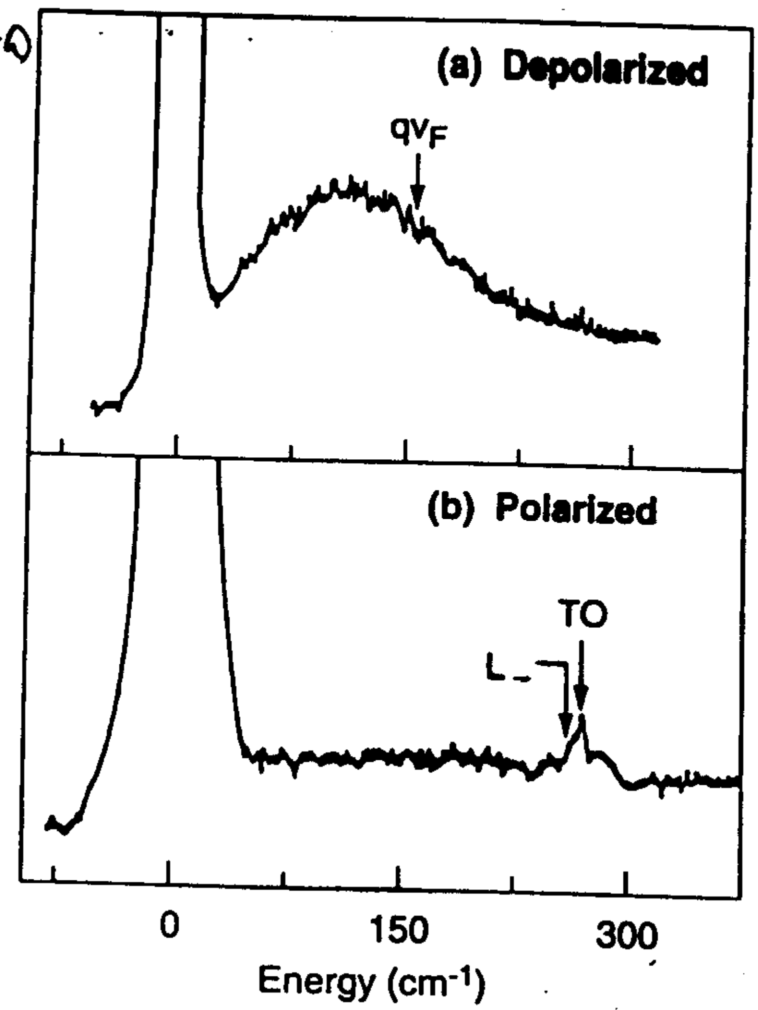

w_i
w_s

$(E_0^{(n)} + \Delta_0)$ gap

n – GaAs $n = 2.0 \times 10^{18}\ cm^{-3}$
T = 10K $\hbar\omega_L = 1915.7$ meV

non-spin flip
ρ excitations
screened by Σ(ε-Δ)





S.P. spin-flip

$e_s \perp e_i$
spin-flip
in
semiconductors
with large
spin-orbit
splitting

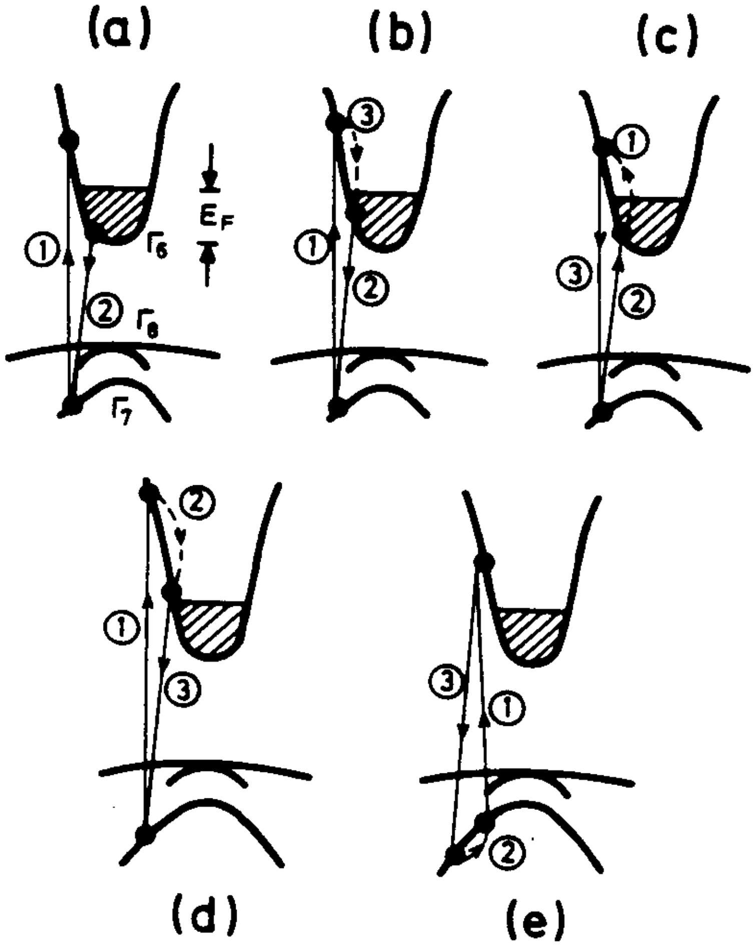


Fig. 1: Diagrams of the various Raman processes discussed in this paper. (a) single particle spin-flip scattering, (b) and (c) electron-phonon collective excitations (charge-density mechanism), (d) and (e) "Fröhlich mechanism". The dashed lines represent the Fröhlich interaction of the longitudinal collective modes. The numbers in circles give the time order of the processes.

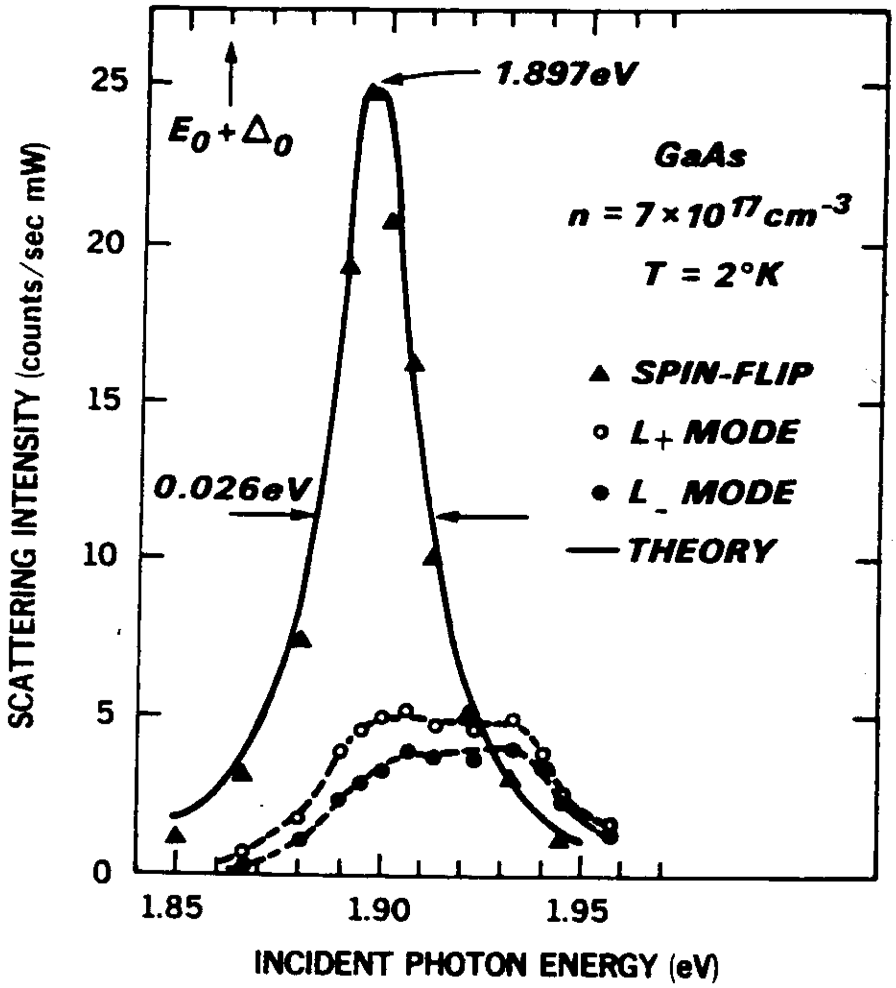


Fig. 2: Scattering intensity as function of incident photon energy of spin-flip single particle excitations at 90 cm^{-1} and the coupled modes at 245 cm^{-1} and 350 cm^{-1}. The full line is the fit with Eq. 3.