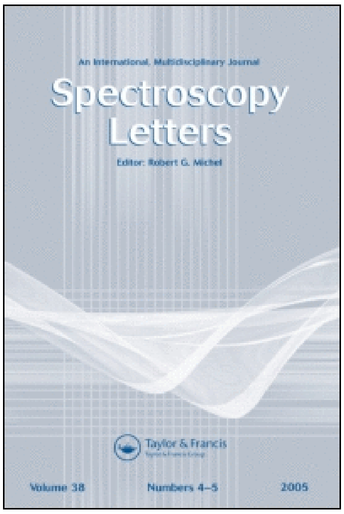

Spectroscopy Letters
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713597299

Kinematical and Dynamical Inelastic Light Scattering in Conducting Crystals

O. Keller[a]
[a] Group of Physics, Institute 7 Aalborg University Centre, Aalborg, Denmark

To cite this Article Keller, O.(1978) 'Kinematical and Dynamical Inelastic Light Scattering in Conducting Crystals', Spectroscopy Letters, 11: 3, 187 — 211

To link to this Article: DOI: 10.1080/00387017808067745

URL: http://dx.doi.org/10.1080/00387017808067745

SPECTROSCOPY LETTERS, 11(3), 187-211 (1978)

KINEMATICAL AND DYNAMICAL INELASTIC LIGHT SCATTERING IN CONDUCTING CRYSTALS *

O. Keller
Group of Physics, Institute 7
Aalborg University Centre, DK-9100 Aalborg, Denmark

Fundamentals of the kinematical quantum theory of inelastic light scattering from electronic excitations in conducting crystals are reviewed. Keeping both the $|\vec{A}|^2$ and $\vec{p}\cdot\vec{A}$ terms in the photon-electron interaction the general expression for the differential scattering cross-section is obtained. Emphasizing opacity effects and anisotropies arising from the Doppler shift the scattering kinematics is analyzed. The scattering from free-carrier density fluctuations is considered briefly. The basic concepts of a recently established phenomenological theory of dynamical light diffraction in opaque crystals are discussed, and some aspects of a predicted phonon induced anomalous transmission of light below the plasma edge in semiconductors are studied.

---

* Presented by invitation of the GPS Chairman at the 51-st Annual Meeting of the Louisiana Academy of Sciences, February 4, 1977, Shreveport, Louisiana, USA.

I. INTRODUCTION

The purpose of this paper is twofold. One is to review some of the basic properties of the quantum theory of kinematical inelastic light scattering from electronic excitations in conducting crystals. The books by Cardona et al.[1], and by Platzman and Wolff[2] are excellent general references for this subject. The other is to present a recently established semi-classical theory of sound induced dynamical light diffraction in semiconductors [3].

In section II the differential scattering cross-section is obtained using time-dependent perturbation theory. The contributions to the cross-section arising from the $|\vec{A}|^2$ and $\vec{p}\cdot\vec{A}$ terms in the photon-electron interaction Hamiltonian are calculated by first- and second-order perturbation expansion, respectively.

The inelastic scattering kinematics in transparent and opaque crystals is considered in section III, and it is shown that anisotropy effects may occur as a consequence of the Doppler shift associated with the scattering event even if frequency - and angular dispersions of the refractive index are negligible.

In section IV we discuss the scattering from free carriers in the limit where the cross terms in the $|\vec{A}|^2$ electron-photon coupling dominate. The general theory is applied to scattering from a noninteracting electron gas.

Section V is devoted to a study of induced dynamical diffraction of light in semi-infinite opaque crystals. Using a quasi-static two-wave interference approximation the complex wave vectors of the optical eigenmodes are determined, and the condition for induced transparency is given. Finally, the theory is applied to the case where the diffraction from a collisionless solid-state plasma dominates.

II. DIFFERENTIAL SCATTERING CROSS-SECTION

The system we are concerned with in this section consists of a radiation field, described by a radiation Hamiltonian $\hat{H}_R$, and a material system, described by a Hamiltonian $\hat{H}_M$. Radiation and matter interact via an interaction Hamiltonian $\hat{H}_{MR}$. In an inelastic light scattering process a quantum of the incident radiation is annihilated and a quantum of the scattered radiation is created. In a Stokes process a collective excitation or a quasi-particle is created whereas an elementary crystal excitation is annihilated in an anti-Stokes event. The anti-Stokes process can occur only if the crystal is initially in an excited state.

In the absence of any interaction between the radiation and matter fields the unperturbed Hamiltonian for the combined system of photons and electrons is given by

$$\hat{H}_0 = \hat{H}_M + \sum_{\vec{k},\mu} \hbar\omega_{\vec{k}\mu} (\hat{a}^{\dagger}_{\vec{k}\mu} \hat{a}_{\vec{k}\mu} + \tfrac{1}{2}) \quad . \tag{1}$$

In the last term, which represents $\hat{H}_R$ in the second-quantization formalism, the creation and destruction operators for photons of wave vector $\vec{k}$, polarization μ, and angular frequency $\omega_{\vec{k}\mu}$ have been denoted by $\hat{a}^{\dagger}_{\vec{k}\mu}$ and $\hat{a}_{\vec{k}\mu}$.

The coupling of non-relativistic electrons, with charge $-e$ and mass m, to a radiation field described by the vector potential $\vec{A}$, can be analyzed by decomposing the interaction Hamiltonian into two terms

$$\hat{H}_{MR} = \hat{H}_A + \hat{H}_{AA} \quad . \tag{2}$$

The quantum-mechanical expression for the term $\hat{H}_A$, which is linear in $\vec{A}$, can be written

$$\hat{H}_A = \frac{e}{m} \sum_{\vec{k},\mu} \left(\frac{\hbar}{2\varepsilon_0 \omega_{\vec{k}\mu} V}\right)^{\frac{1}{2}} \left[\hat{\vec{P}}(-\vec{k}) \cdot \vec{e}_{\vec{k}\mu} \hat{a}_{\vec{k}\mu} + \text{H.C.}\right], \tag{3}$$

where $\vec{e}_{\vec{k}\mu}$ is the unit polarization vector for a transverse photon $\vec{k},\mu$. The normalization volume is V. The term $\hat{H}_{AA}$, which is quadratic in the vector potential, is given by

$$\hat{H}_{AA} = \frac{e^2}{2m} \sum_{\vec{k}_1,\mu_1} \sum_{\vec{k}_2,\mu_2} \left(\frac{\hbar}{2\varepsilon_0 \omega_{\vec{k}_1\mu_1} V}\right)^{\frac{1}{2}} \left(\frac{\hbar}{2\varepsilon_0 \omega_{\vec{k}_2\mu_2} V}\right)^{\frac{1}{2}} \times$$

$$\left[\hat{N}(-\vec{k}_1-\vec{k}_2)\vec{e}_{\vec{k}_1\mu_1} \cdot \vec{e}_{\vec{k}_2\mu_2} \hat{a}_{\vec{k}_1\mu_1} \hat{a}_{\vec{k}_2\mu_2} + \right.$$

$$\left. \hat{N}(-\vec{k}_1+\vec{k}_2)\vec{e}_{\vec{k}_1\mu_1} \cdot \vec{e}^{\,*}_{\vec{k}_2\mu_2} \hat{a}_{\vec{k}_1\mu_1} \hat{a}^{\dagger}_{\vec{k}_2\mu_2} + \text{H.C.}\right] . \tag{4}$$

In Eqs. (3) and (4) the Fourier transform of the many-particle number and momentum operators of the electrons, i.e.

$$\hat{N}(\vec{k}) = \sum_j e^{-i\vec{k}\cdot\vec{r}_j} = \int e^{-i\vec{k}\cdot\vec{r}} \sum_j \delta(\vec{r}-\vec{r}_j) d^3r =$$

$$\int e^{-i\vec{k}\cdot\vec{r}} N(\vec{r}) d^3r \quad , \tag{5}$$

and analogous

$$\hat{\vec{P}}(\vec{k}) = \sum_j e^{-i\vec{k}\cdot\vec{r}_j} \hat{\vec{p}}_j \quad , \tag{6}$$

have been introduced; $\vec{r}_j$ and $\hat{\vec{p}}_j$ being the position and momentum operators of the jth electron. The normalization condition used for the vector potential is such that the observable energy flux density of photons in the mode $\vec{k}\mu$ becomes

$$\vec{I}_{\vec{k}\mu} = \frac{1}{V}c^2\hbar\vec{k}n_{\vec{k}\mu} \tag{7}$$

i.e. with the zero-point contribution removed, the expectation value of the Poynting vector operator for the mode. The number of photons in the mode has been denoted by $n_{\vec{k}\mu}$.

Since light scattering is a two-photon process in which the scattering medium through the electron-radiation interaction destroys the incident photon and creates the scattered photon it follows from Eqs. (3) and (4) that a time-dependent perturbation calculation of the rate of scattering from an initial state $|I\rangle$ to a final state $|F\rangle$ of the unperturbed system described by the Hamiltonian $\hat{H}_0$ has to be carried out to first order in $\hat{H}_{AA}$ (The Fermi golden rule) and to second order in $\hat{H}_A$. The transition probability per unit time consistent with a given final energy for the scattered particle is thus [4]

$$\frac{1}{\tau_{I\to F}} = \frac{2\pi}{\hbar}\sum_F \left| \langle F|\hat{H}_{AA}|I\rangle + \sum_L \frac{\langle F|\hat{H}_A|L\rangle\langle L|\hat{H}_A|I\rangle}{E_I-E_L} \right|^2 \delta(E_I-E_F). \tag{8}$$

The states $|L\rangle$ which occur in the second term of Eq. (8) are the virtual intermediate states for the transition. The energy eigenvalues of the unperturbed states have been denoted E_I, E_L, and E_F.

By combining Eqs. (3), (4), and (8) it appears that the transition rate from an initial photon state $(\mu_1, \vec{k}_1, \omega_{\vec{k}_1\mu_1})$ to a final state $(\mu_2, \vec{k}_2, \omega_{\vec{k}_2\mu_2})$ (in the following often for shortness denoted $(1, \omega_1)$ and $(2, \omega_2)$) can be written more explicitly as

$$\frac{1}{\tau_{1\to2}} = \frac{2\pi}{\hbar}\sum_f \Big| \langle n-1,1,f|\hat{H}_{AA}|n,0,i\rangle +$$

$$\sum_\ell \Big[\frac{\langle n-1,1,f|\hat{H}_A|n-1,0,\ell\rangle\langle n-1,0,\ell|\hat{H}_A|n,0,i\rangle}{E_i-E_\ell+\hbar\omega_1} +$$

$$\frac{\langle n-1,1,f|\hat{H}_A|n,1,\ell\rangle\langle n,1,\ell|\hat{H}_A|n,0,i\rangle}{E_i-E_\ell-\hbar\omega_2} \Big]\Big|^2 \delta(E_i-E_f+\hbar\omega_1-\hbar\omega_2). \qquad (9)$$

The first and second entrities in the bra and kets for the total final and initial states refer in the occupation-number formalism to the initial and final photon states. The energies of the incident and scattered photons are $h\omega_1$ and $h\omega_2$, respectively. The exact initial, intermediate, and final many-body states of the crystal are $|i\rangle$, $|\ell\rangle$, and $|f\rangle$; the energies of the states being E_i, E_ℓ, and E_f. The overall energy conservation in the scattering process is expressed in terms of the delta function.

The three contributions to the scattering matrix element are shown graphically in Fig. 1. Diagram (a) corresponds, in accordance with the second order character of the Hamiltonian $\hat{H}_{AA}$, to a simultaneous absorption and emission of photons 1 and 2. In diagram (b) the material system absorbs in a virtual process a photon of angular frequency ω_1, and then emits a photon of frequency ω_2, and in diagram (c) the crystal system emits ω_2 first, and then absorbs ω_1.

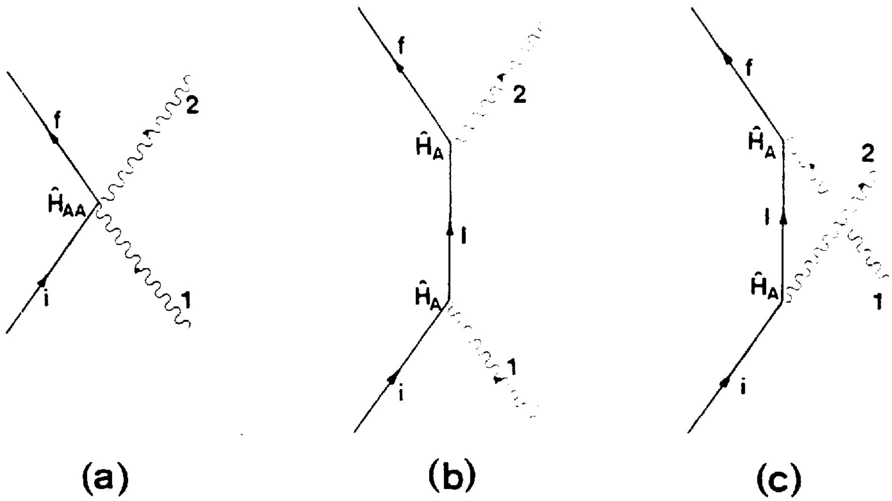

Fig. 1 Feynman diagrams representing the three possible contributions to the first-order inelastic light scattering matrix element. Diagram (a) describes the first-order interaction via the Hamiltonian $\hat{H}_{AA}$, and diagrams (b) and (c) the two types of second-order interaction arising from the operator $\hat{H}_A$.

The rate of energy removal from the incident beam is $\sum_2 \hbar\omega_2/\tau_{1\to 2}$ where the summation is over possible scattered photon states μ, $\vec{k}_2$. Since the incident light intensity is $c\hbar\omega_1 n_1/V$ the total *power* scattering cross-section is given by

$$\sigma = \frac{V}{c}\,\frac{\sum\limits_{\vec{k}_2,\mu_2} \omega_{\vec{k}_2\mu_2}/\tau_{\vec{k}_1\mu_1\to\vec{k}_2\mu_2}}{\omega_{\vec{k}_1\mu_1}\, n_{\vec{k}_1\mu_1}} \tag{10}$$

The differential scattering cross-section is obtained from Eq. (10) by converting the summation over $\vec{k}_2\mu_2$ to an integration,

$$\sum_{\vec{k}_2\mu_2} \rightarrow \frac{V}{(2\pi)^3}\iint \frac{\omega^2_{\vec{k}_2\mu_2}}{c^3}\, d\omega_{\vec{k}_2\mu_2}\, d\Omega \quad , \tag{11}$$

where $d\Omega$ is an element of solid angle around $\vec{k}_2$.

By combining Eqs. (3), (4), (9), (10), and (11) and utilizing that the only non-vanishing matrix elements of $\hat{a}_{\vec{k}\mu}$ and $\hat{a}^{\dagger}_{\vec{k}\mu}$ are

$$\langle n_{\vec{k}\mu}|\hat{a}^{\dagger}_{\vec{k}\mu}|n_{\vec{k}\mu}-1\rangle = \langle n_{\vec{k}\mu}-1|\hat{a}_{\vec{k}\mu}|n_{\vec{k}\mu}\rangle = (n_{\vec{k}\mu})^{\frac{1}{2}} \quad , \tag{12}$$

the differential *photon* scattering cross-section, which is ω_1/ω_2 times the power scattering cross-section, can be written

$$\frac{d^2\sigma}{d\omega_2 d\Omega} = \left(\frac{e^2}{4\pi\varepsilon_0 mc^2}\right)^2 \left(\frac{\omega_2}{\omega_1}\right) \sum_{i,f} P(E_i)\,|\vec{e}_2\cdot\delta\overleftrightarrow{\chi}^{i,f}\cdot\vec{e}_1|^2\ \delta\!\left(\frac{E_i-E_f}{\hbar}+\omega_1-\omega_2\right), \tag{13}$$

where $\delta\overleftrightarrow{\chi}^{i,f}$ is a second-rank tensor with components $\langle f|\delta\chi_{ab}|i\rangle$ given by

$$\langle f|\delta\chi_{ab}|i\rangle = \langle f|\hat{N}(-\vec{k}_1+\vec{k}_2)|i\rangle\delta_{ab} +$$

$$\frac{1}{m}\sum_{\ell}\left[\frac{\langle f|P_a(\vec{k}_2)|\ell\rangle\langle\ell|P_b(-\vec{k}_1)|i\rangle}{E_i+\hbar\omega_1-E_\ell} + \right.$$

$$\left.\frac{\langle f|P_b(-\vec{k}_1)|\ell\rangle\langle\ell|P_a(\vec{k}_2)|i\rangle}{E_i-\hbar\omega_2-E_\ell}\right] \quad . \tag{14}$$

The matrix element in Eq. (14) is proportional to the ab component of the transition electric susceptibility tensor. As indicated by the Kronecker delta δ_{ab}, the first term on the and temporal fluctuations in the electronic contribution to the electric susceptibility, i.e.

$$\delta\chi_{12}(\vec{k},t) = \frac{1}{V}\int_V \exp(-i\vec{k}\cdot\vec{r})\,\delta\chi_{12}(\vec{r},t)\,d^3r \quad , \tag{23}$$

where the fluctuation due to the elementary excitation $(\Omega_{\vec{q}},\vec{q})$ is given by

$$\delta\chi_{12}(\vec{r},t) = \delta\chi_{12}(\vec{q})\exp\left[i(\vec{q}\cdot\vec{r}-\Omega_{\vec{q}}t)\right]+c.c. \quad . \tag{24}$$

In opaque crystals the optical wave vectors are complex i.e. $\vec{k}_1 = \mathrm{Re}\vec{k}_1+i\mathrm{Im}\vec{k}_1$ and $\vec{k}_2 = \mathrm{Re}\vec{k}_2+i\mathrm{Im}\vec{k}_2$.

If the scattering volume is a rectangular parallellepiped $V = \pi_i\, a_i$ it follows from the treatment in section II, and from Eqs. (23) and (24) that the scattering cross-section in an opaque crystal is proportional to

$$\frac{d^2\sigma}{d\omega_2 d\Omega} \propto \prod_{i=1}^{3} \left\{\left[q_i-(\mathrm{Re}k)_i\right]^2+\left[(\mathrm{Im}k_1)_i+(\mathrm{Im}k_2)_i\right]^2\right\}^{-1} \quad , \tag{25}$$

in the limit where the linear extensions of the scattering volume are large in comparison with the penetration depths of the radiation i.e. $a_i(\mathrm{Im}k_1)_i$, $a_i(\mathrm{Im}k_2)_i \gg 1$. Thus, it is realized that the inelastic scattering in opaque media is due to excitations having a range of wave vectors

$$\Delta\vec{q} = \mathrm{Im}\vec{k}_1+\mathrm{Im}\vec{k}_2 \tag{26}$$

around

$$\vec{q} = \mathrm{Re}\vec{k}_1-\mathrm{Re}\vec{k}_2 \tag{27}$$

The ratio between the magnitudes of the incident and scattered wave vectors is given by

$$\frac{k_2}{k_1} = \frac{n_2(\omega_2,\vec{e}_2)}{n_1(\omega_1,\vec{e}_1)}\left[1 \mp \frac{\Omega_{\vec{q}}}{\omega_1}\right] \quad , \tag{17}$$

where n_1 and n_2 are the refractive indices of the crystal for the incident and scattered light beams.

According to Eq. (17) deviations from the isosceles pseudomentum triangle, which is well known from elastic Bragg diffraction, can occur if the crystal is optically anisotropic or if light frequency dispersion effects are of importance. However, it also appears from the last factor in Eq. (17) that the normal Bragg law is violated "directly" as a consequence of the Doppler shift associated with an inelastic scattering process. Implications for the scattering kinematics arising from this fact, which seem to have been almost overlooked in the litterature, are discussed shortly below. Thus, for an isotropic solid where the frequency dispersion of the refractive index can be neglected, one obtains by combining Eqs. (15) and (16) the following expressions for the Bragg angles (θ_1,θ_2) of the incident and scattered light

$$\sin\theta_1 = \frac{\Omega_{\vec{q}}}{2\omega_1}\left[\frac{1}{\beta} - \beta\right] \pm \beta \quad , \tag{18}$$

and

$$\sin\theta_2 = \frac{1}{1 + \frac{\Omega_{\vec{q}}}{\omega_1}}\left\{\frac{\Omega_{\vec{q}}}{2\omega_1}\left[\frac{1}{\beta} - \beta\right] \mp \beta\right\} \quad , \tag{19}$$

where we have introduced the ratio

$$\beta = \frac{n\Omega_{\vec{q}}}{cq} \tag{20}$$

between the phase velocities of the crystal excitation and the light. Restricting the analysis to optical energies which are large compared to the crystal excitation energies i.e. $\Omega_{\vec{q}} << \omega_1$ Eqs. (18) and (19) are reduced to

$$\sin\theta_1 = \frac{\Omega_{\vec{q}}}{2\omega_1} \frac{1}{\beta} \pm \beta \quad , \tag{21}$$

and

$$\sin\theta_2 = \frac{\Omega_{\vec{q}}}{2\omega_1} \frac{1}{\beta} \mp \beta \quad . \tag{22}$$

Let us apply the results obtained in Eqs. (21) and (22) to scattering from (i) optical phonons characterized by the simple dispersion relation $\Omega_{\vec{q}} = \Omega_0$, independent of q, and (ii) acoustic phonons showing no dispersion i.e. $\Omega_{\vec{q}} = V_p q$, where the acoustic phase velocity V_p is assumed to be independent of $\vec{q}$. For clearness, only the Stokes process is considered below. In case (i), there occur a minimum value of q obtained in forward scattering and given by $q_{min} \simeq n\Omega_{\vec{q}}/c$ as indicated in Fig. 2. The optical wave vectors are in this limit parallel to $\vec{q}_{min}$. The maximum value of q is obtained in backscattering and given by $q_{max} \simeq 2n\omega_1/c$. Inbetween these limits there is a minimum in $\sin\theta_1$ at $q = (n/c)(2\omega_1\Omega_0)^{\frac{1}{2}}$. The quantity $\sin\theta_2$ is a monotonically increasing function of q, which has a zero coincident with the minimum in $\sin\theta_1$. In case (ii) $\sin\theta_1$ and $\sin\theta_2$ increase both linearly towards $q_{max} \simeq 2n\omega_1/c$ since the ratio between the acoustical and optical phase velocities is much smaller than unity. For $q \to 0$ one obtains $\sin\theta_1 \simeq -\sin\theta_2 \to \sin\theta_0 = V_p/(c/n) << 1$, which shows that the scattering is forward but with the acoustical and optical wave vectors almost perpendicular to each other (see Fig. 3).

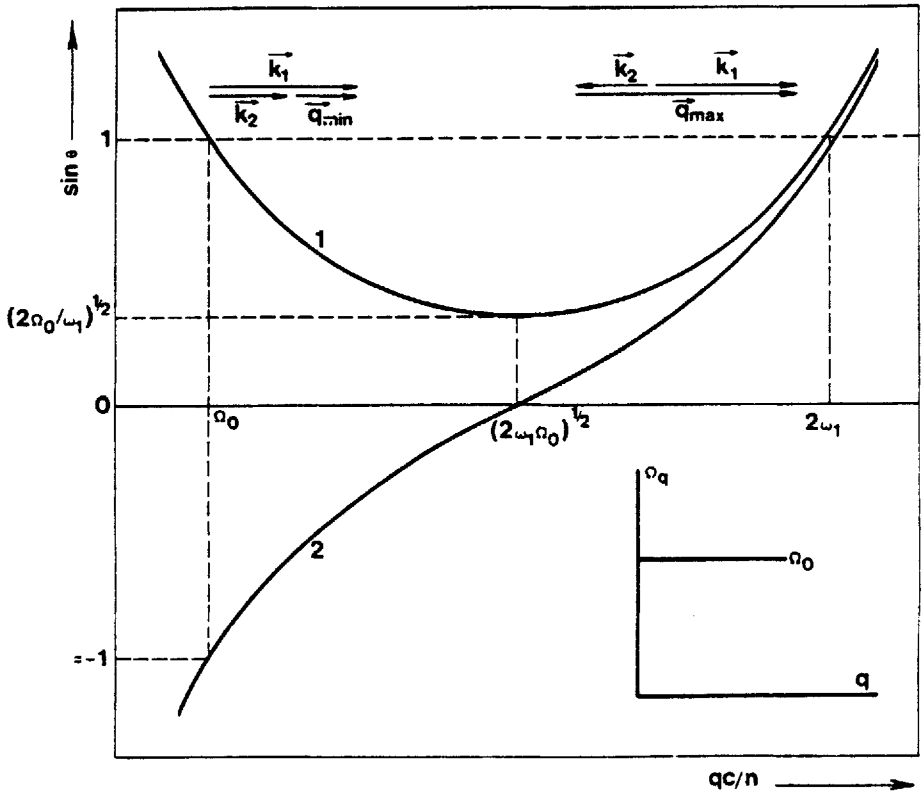

Fig. 2 Sine of the Bragg angles of the incident (1) and scattered (2) photons as a function of the reduced optical phonon wave vector for a Stokes process. The dispersion relation of the optical phonon, and the scattering kinematics for the limiting phonon wave vectors are shown in the inserts. The kinematics is highly anisotropic for phonons in the long wavelength region.

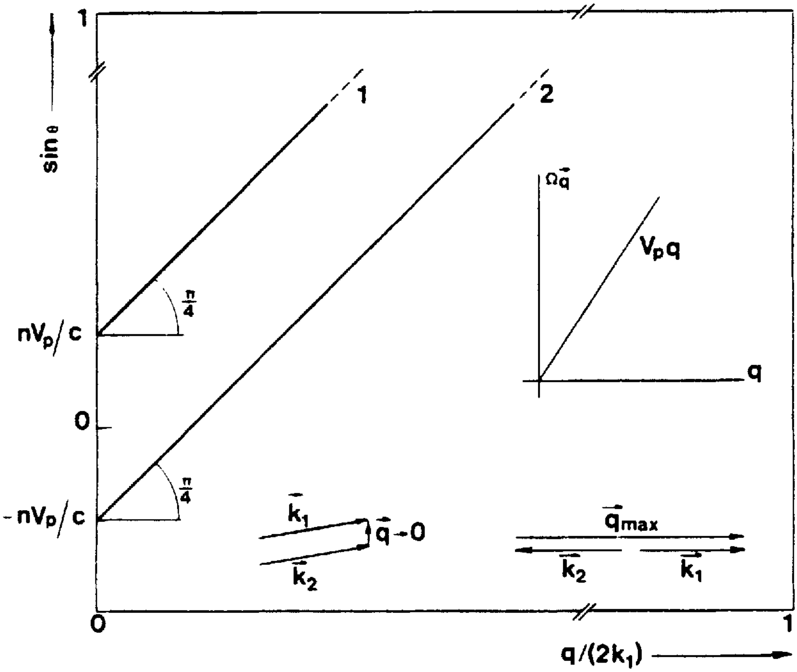

Fig. 3 Sine of the Bragg angles of the incident (1) and scattered (2) photons as a function of the normalized acoustic phonon wave vector for a Stokes process. The vertical separation of the two linear curves is $2nV_p/c \ll 1$. The inserts show the acoustic phonon dispersion relation, and the scattering kinematics for the limiting phonon wave vectors.

To analyze the scattering kinematics in opaque crystals one makes use of the fact (see section IV for the details) that the scattering cross-section is proportional to $\langle i|\delta\hat{\chi}^{\dagger}_{12}(\vec{k},t)\delta\hat{\chi}_{12}(\vec{k},0)|i\rangle$, where $\vec{k} = \vec{k}_1 - \vec{k}_2$ is the scattering wave vector, and $\delta\chi_{12}(\vec{k},t) = \vec{e}_2\cdot\delta\overleftrightarrow{\chi}^{i,f}(\vec{k},t)\cdot\vec{e}_1$. The operator $\delta\hat{\chi}_{12}(\vec{k},t)$ is proportional to the Fourier transform of the spatial

IV. SCATTERING FROM FREE CARRIERS

The summation over final states f of the many-body system appearing in Eq. (13) can be carried out formally by using the Dirac delta-function representation

$$\delta\left(\frac{E_i-E_f}{\hbar}+\omega_1-\omega_2\right)=\frac{1}{2\pi}\lim_{T\to\infty}\int_{-T}^{T}\exp\left[i\left(\frac{E_i-E_f}{\hbar}+\omega_1-\omega_2\right)t\right]dt, \tag{28}$$

the closure theorem

$$\sum_f |f\rangle\langle f| = 1 \quad , \tag{29}$$

and the relation

$$\langle i|\hat{O}(t)|f\rangle = e^{i(E_i-E_f)t/\hbar}\langle i|\hat{O}(0)|f\rangle \quad , \tag{30}$$

where the perturbation operator $\hat{O}$ in the Heisenberg representation is given by

$$\hat{O}(t) = e^{i\hat{H}_M t/\hbar}\,\hat{O}(0)\,e^{-i\hat{H}_M t/\hbar} \quad . \tag{31}$$

As a result of this procedure one obtains

$$\frac{d^2\sigma}{d\omega_2 d\Omega} = r_0^2\left(\frac{\omega_2}{\omega_1}\right)\sum_i P(E_i)\int_{-\infty}^{\infty} e^{i(\omega_1-\omega_2)t}\langle i|\delta\hat{\chi}_{12}^{\dagger}(\vec{k},t)\,\delta\hat{\chi}_{12}(\vec{k},0)|i\rangle\frac{dt}{2\pi}, \tag{32}$$

where $\delta\hat{\chi}_{12}(\vec{k},t) = \vec{e}_2\cdot\delta\hat{\chi}^{i,f}(\vec{k},t)\cdot\vec{e}_1$ is proportional to the transition susceptibility operator in the Heisenberg representation.

When the optical energies are large compared to important excitation energies of the many-body system the scattering from the solid state plasma is usually dominated by the cross terms in the $\hat{H}_{AA}$ electron-photon coupling. Neglecting the second term in Eq. (8), the scattering cross-section may be written as

$$\frac{d^2\sigma}{d\omega_2 d\Omega} = V^2 r_0^2 \left(\frac{\omega_2}{\omega_1}\right) (\vec{e}_2 \cdot \vec{e}_1)^2 \sum_i P(E_i) \int_{-\infty}^{\infty} e^{i\Omega t} \langle i | \hat{n}^\dagger(-\vec{Q},t) \hat{n}(-\vec{Q},0) | i \rangle \frac{dt}{2\pi}, \tag{33}$$

where $\hat{n}(\vec{Q},t) = \hat{N}(\vec{Q},t)/V$ is the Fourier transform of the electron density operator.

If the crystal is initially in thermal equilibrium the scattering cross-section per unit volume (V = 1) takes the form

$$\frac{d^2\sigma}{d\omega_2 d\Omega} = r_0^2 \left(\frac{\omega_2}{\omega_1}\right) (\vec{e}_2 \cdot \vec{e}_1)^2 S(\vec{Q},\Omega) \quad , \tag{34}$$

where the quantity

$$S(\vec{Q},\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\Omega t} \langle i | \hat{n}^\dagger(-\vec{Q},t) \hat{n}(-\vec{Q},0) | i \rangle_T dt \quad , \tag{35}$$

is the so-called dynamical structure factor, and where $\langle \ldots \rangle_T$ denotes the thermal ensemble average over the crystal initial states. The fact that the dynamical structure factor is the space-time Fourier transform of the electron density-density correlation function i.e.

$$S(\vec{Q},\Omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{i\Omega t} \int_{\vec{r}} \int_{\vec{r}'} e^{-i\vec{Q}\cdot(\vec{r}-\vec{r}')} \langle i | \hat{n}(\vec{r},t) \hat{n}(\vec{r}',0) | i \rangle_T d^3r d^3r' dt, \tag{36}$$

indicates that the electron density fluctuations are responsible for the scattering from the plasma.

In the simple case of a noninteracting electron gas the dynamical structure factor $S_0(\vec{Q},\Omega)$ can easily be evaluated. In second quantized notation, one has

$$\hat{n}(\vec{Q},0) = \sum_{\vec{K}} \hat{C}^\dagger_{\vec{K}+\vec{Q}} \hat{C}_{\vec{K}} \tag{37}$$

where $\hat{C}^{\dagger}_{\vec{K}}$ and $\hat{C}_{\vec{K}}$ are the usual creation and annihilation operators. Combining Eq. (37), the Hamiltonian

$$\hat{H}_M = \sum_{\vec{K}} \hat{C}^{\dagger}_{\vec{K}} \hat{C}_{\vec{K}} \frac{\hbar^2 K^2}{2m} \tag{38}$$

and the Fermion commutation rule

$$\left[\hat{C}_{\vec{K}}, \hat{C}^{\dagger}_{\vec{L}}\right]_+ = \hat{C}_{\vec{K}} \hat{C}^{\dagger}_{\vec{L}} + \hat{C}^{\dagger}_{\vec{L}} \hat{C}_{\vec{K}} = \delta(\vec{L}-\vec{K}) \quad , \tag{39}$$

one can show that

$$S_0(\vec{Q},\Omega) = \sum_{\vec{K}} \left\{ f_0(E_{\vec{K}}) \left[1-f_0(E_{\vec{K}+\vec{Q}})\right] \delta\left(\frac{E_{\vec{K}}-E_{\vec{K}+\vec{Q}}}{\hbar} + \Omega\right) \right\} \quad , \tag{40}$$

where

$$f_0(E_{\vec{K}}) = \langle i | \hat{C}^{\dagger}_{\vec{K}} \hat{C}_{\vec{K}} | i \rangle_T \quad , \tag{41}$$

is the Fermi-Dirac distribution function, and $E_{\vec{K}} = \frac{\hbar^2 K^2}{2m}$ the energy of the plane electron wave having a wave vector $\vec{K}$. It follows directly from Eq. (40) that $S_0(\vec{Q},\Omega)$ represents the sum of all possible scattering events in which an electron goes from a filled state $\vec{K}$ to an empty state $\vec{K}+\vec{Q}$ with conservation of energy and momentum between the target and the incident photon.

V. DYNAMICAL DIFFRACTION

When the crystal is opaque to the incident and scattered light interference effects among the plane wave components of the optical wave field must be considered by replacing the usual kinematical theory of inelastic scattering by a dynamical one.

In this section some fundamentals of a recently established phenomenological theory, which is based on a two-wave interference approximation, are outlined. It is assumed that (I) the crystal is semi-infinite, (II) the wave vector of the dielectric disturbance is parallel to the surface, (III) the scattering plane is perpendicular to the boundary plane, (IV) only bulk scattering effects are of importance, and (V) a quasi-static approximation ($\omega_1 \gg \Omega_{\vec{q}}$) is valid.

A. Boundary effects

Limiting the analysis to isotropic scattering geometries one finds that the real wave vectors of the incident ($\vec{k}_{01}$) and scattered ($\vec{k}_{02}$) optical waves outside the crystal are related by the equation

$$\vec{k}_{02} = -(\vec{k}_{01} \pm \vec{q}). \tag{42}$$

Inside the crystal the incident and scattered waves are inhomogeneous. Elementary considerations based on Maxwell's equations and Bragg's law show that the real parts of the "inside" wave vectors fulfil the condition

$$\mathrm{Re}\vec{k}_2 = \mathrm{Re}\vec{k}_1 \pm \vec{q} \tag{43}$$

and the imaginary parts the relation

$$\mathrm{Im}\vec{k}_1 = \mathrm{Im}\vec{k}_2 = \gamma\vec{n} \tag{44}$$

where $\vec{n}$ is a unit vector perpendicular to the surface, and γ is the amplitude attenuation coefficient of the modes.

B. Wave field

The wave field in the crystal is obtained by solving the inhomogeneous time-independent wave equation for the total field $\vec{E}(\vec{r},\omega)$ neglecting the Doppler shifts ($\omega = \omega_2 \simeq \omega_1$)

$$\vec{\nabla}\,\vec{\nabla}\cdot\vec{E}(\vec{r},\omega)-\nabla^2\vec{E}(\vec{r},\omega) = \left(\frac{\omega}{c}\right)^2\left[\overleftrightarrow{\varepsilon}_0 + \overleftrightarrow{\varepsilon}(\vec{r},\omega)\right]\cdot\vec{E}(\vec{r},\omega), \qquad (45)$$

where $\overleftrightarrow{\varepsilon}_0$ is the unperturbed relative dielectric tensor, and $\overleftrightarrow{\varepsilon}(\vec{r},\omega)$ is the time-independent part of the harmonic plane-wave component $(\Omega_{\vec{q}},\vec{q})$ of the dielectric disturbance. The amplitude will be named $\overleftrightarrow{\varepsilon}_{\vec{q}}$. The gradient operator has been denoted by $\vec{\nabla}$.

Restricting the discussion to the interesting case where the incident and scattered waves are polarized perpendicular to the scattering plane, one obtains in a two-wave interference approximation

$$\vec{E}(\vec{r},\omega) = \vec{E}_0 \exp\left(i\,\frac{\phi_{\vec{q}}}{2}\right) \times$$

$$\Big\{\exp\left[(ik_\perp^+ - \gamma_\perp^+)\vec{n}\cdot\vec{r}\right]\cos\tfrac{1}{2}(\vec{q}\cdot\vec{r} + \phi_{\vec{q}}) -$$

$$i\exp\left[(ik_\perp^- - \gamma_\perp^-)\vec{n}\cdot\vec{r}\right]\sin\tfrac{1}{2}(\vec{q}\cdot\vec{r} + \phi_{\vec{q}})\Big\} \quad , \qquad (46)$$

where the real and imaginary parts of the wave vector components of the two eigenmodes (+,−) perpendicular to the surface are given by

$$k_\perp^\pm = \frac{\omega}{c}\left\{\tfrac{1}{2}\left[\sqrt{\left[\mathrm{Re}\tilde{\varepsilon}_0 \pm |\tilde{\varepsilon}_{\vec{q}}| - \left(\frac{qc}{2\omega}\right)^2\right]^2 + (\mathrm{Im}\tilde{\varepsilon}_0)^2}\right.\right.$$

$$\left.\left. + \mathrm{Re}\tilde{\varepsilon}_0 \pm |\tilde{\varepsilon}_{\vec{q}}| - \left(\frac{qc}{2\omega}\right)^2\right]\right\}^{\frac{1}{2}} \quad , \qquad (47)$$

and

$$\gamma_{\perp}^{\pm} = \frac{\omega}{c} \left\{ \tfrac{1}{2} \left[\sqrt{\left[\mathrm{Re}\tilde{\varepsilon}_0 \pm |\tilde{\varepsilon}_{\vec{q}}| - \left(\frac{qc}{2\omega}\right)^2 \right]^2 + (\mathrm{Im}\tilde{\varepsilon}_0)^2} - \mathrm{Re}\tilde{\varepsilon}_0 \mp |\tilde{\varepsilon}_{\vec{q}}| + \left(\frac{qc}{2\omega}\right)^2 \right] \right\}^{\frac{1}{2}} \quad (48)$$

In the above equations the appropriate components of the complex dielectric tensor have been denoted by a tilde.

The components of the wave vectors of the plus and minus sign modes parallel to the surface are equal and real, and in magnitude given by

$$k_{\parallel}^{\pm} = \frac{q}{2} \quad (49)$$

The dynamical scattering kinematics is shown schematically in Fig. 4.

The phase factor $\phi_{\vec{q}}$, is for the anti-Stokes process given by

$$\mathrm{tg}\phi_{\vec{q}} = \frac{\mathrm{Im}\tilde{\varepsilon}_{\vec{q}}}{\mathrm{Re}\tilde{\varepsilon}_{\vec{q}}} \quad (50)$$

and for the Stokes process by $\phi_{-\vec{q}} = -\phi_{\vec{q}}$. The interpretation of the optical wave pattern in the crystal, as given by Eq. (46), is straightforward.

According to Eq. (48) the condition for highly transparency of the material is

$$0 < \frac{|\mathrm{Im}\tilde{\varepsilon}_0|}{\mathrm{Re}\tilde{\varepsilon}_0 \pm |\tilde{\varepsilon}_{\vec{q}}| - \left(\frac{qc}{2\omega}\right)^2} \ll 1 \quad (51)$$

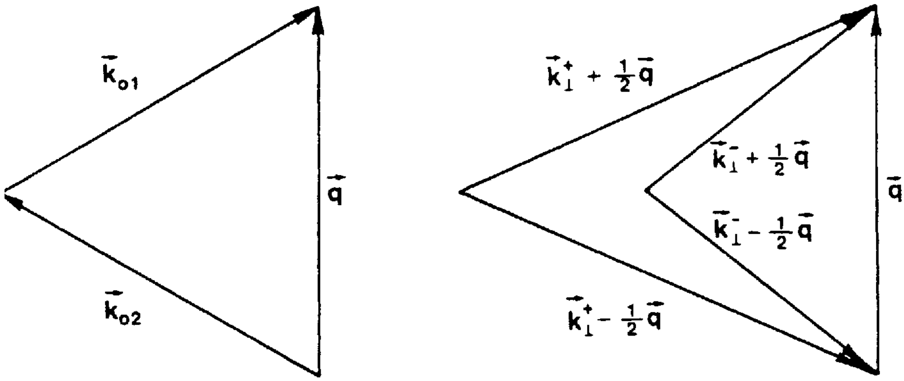

Fig. 4 Quasi-elastic dynamical light diffraction kinematics for the two optical eigenmodes outside and inside the crystal.

It should be noticed that the optical properties of a dielectric perturbed crystal can be obtained from the unperturbed case by making the replacement

$$\mathrm{Re}\tilde{\varepsilon}_0 \rightarrow \mathrm{Re}\tilde{\varepsilon}_0 \pm |\tilde{\varepsilon}_q| - \left(\frac{qc}{2\omega}\right)^2 \tag{52}$$

C. Bunched collisionless plasma

In the following the basic formulation of dynamical diffraction given in the preceding subsections is applied to the case where the scattering from free-carrier density waves dominates. It is assumed that the solid-state plasma is collisionless.

In the long wavelength limit the unperturbed dielectric constant is given by

$$\tilde{\varepsilon}_0 = \tilde{\varepsilon}_0^L\left[1-\left(\frac{\tilde{\omega}_p}{\omega}\right)^2\right] \tag{53}$$

where $\tilde{\varepsilon}_0^L$ is the lattice contribution to the appropriate dielectric constant, and $\tilde{\omega}_p$ is the angular plasma frequency. It is assumed that $\tilde{\varepsilon}_0^L$ is real. The perturbed dielectric constant has an amplitude [5]

$$\tilde{\varepsilon}_{\vec{q}} = -\tilde{\varepsilon}_0^L\left(\frac{\tilde{\omega}_p}{\omega}\right)^2\tilde{\Delta}_{\vec{q}} \quad , \tag{54}$$

where $\tilde{\Delta}_{\vec{q}}$ is the ratio between the amplitude of the space charge density wave and the equilibrium density. In semiconductors the bunching of the free carriers can be obtained via piezoelectric coupling or deformation potential coupling to elastic waves.[6]

Introducing the dynamical plasma frequencies $\tilde{\omega}_{dyn}^{\pm}$ of the forward diffracted eigenmodes

$$\tilde{\omega}_{dyn}^{\pm} = \left[\tilde{\omega}_p^2(1 \mp |\tilde{\Delta}_{\vec{q}}|) + \left(\frac{qc}{2(\tilde{\varepsilon}_0^L)^{\frac{1}{2}}}\right)^2\right]^{\frac{1}{2}} \quad , \tag{55}$$

it follows by combining Eqs. (47), (48), (53), and (54) that the electromagnetic modes propagate almost undamped ($\gamma_\perp^{\pm} \simeq 0$) in the regions $\omega \geq \tilde{\omega}_{dyn}^{\pm}$. The dispersion relations of the modes have the form (see Fig. 5)

$$k_\perp^{\pm} = \frac{\left[\omega^2 - (\tilde{\omega}_{dyn}^{\pm})^2\right]^{\frac{1}{2}}}{c_\infty} \quad , \tag{56}$$

where $c_\infty = c/(\tilde{\varepsilon}_0^L)^{\frac{1}{2}}$ is the "high frequency" velocity of light. In the absorbing regions $\omega \leq \tilde{\omega}_{dyn}^{\pm}$, where $k_\perp^{\pm} \simeq 0$ one obtains

$$\gamma_\perp^{\pm} = \frac{\left[(\tilde{\omega}_{dyn}^{\pm})^2 - \omega^2\right]^{\frac{1}{2}}}{c_\infty} \quad . \tag{57}$$

The splitting of the dispersion relation at zero wave vector is given by

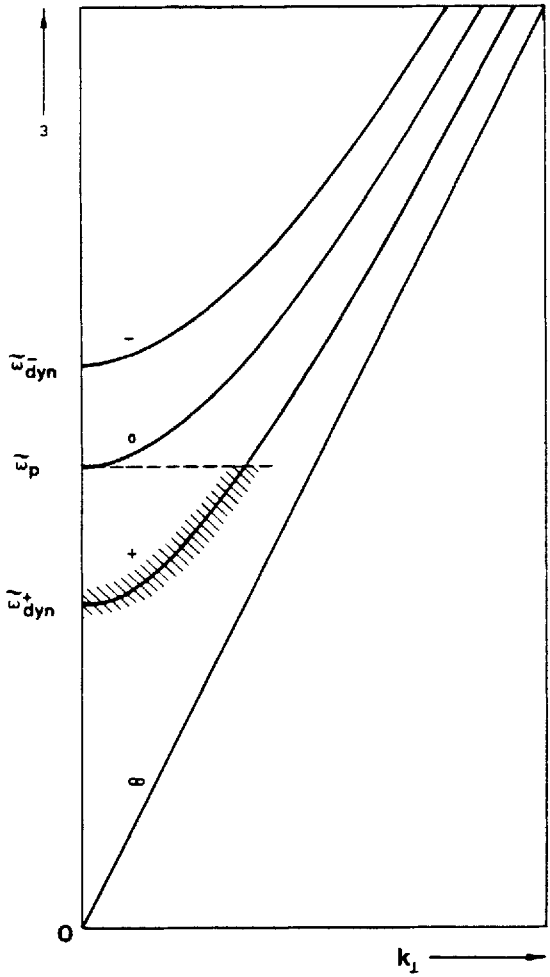

Fig. 5 Dispersion relations for dynamically diffracted (+,-) and undiffracted (0) long wavelength electromagnetic eigenmodes in a collisionless solid-state plasma. Neglecting band structure effects the curves approach a common nondispersive $\omega(\vec{k})$-relation (∞) at high frequencies. The shaded part of the lower branch shows the region of anomalous transmission.

$$(\tilde{\omega}^{-}_{dyn})^2-(\tilde{\omega}^{+}_{dyn})^2 = 2|\tilde{\Delta}_{\vec{q}}|\tilde{\omega}^2_p \quad (58)$$

The minus sign mode suffers enchanced absorption since $\tilde{\omega}_p < \tilde{\omega}^{-}_{dyn}$, whereas the plus sign mode is subjected to a smaller damping than the unperturbed mode. This leads to anomalous transmission of electromagnetic waves below the plasma edge if $\tilde{\omega}^{+}_{dyn} < \tilde{\omega}_p$, a condition which can be expressed by the important inequality

$$|\tilde{\Delta}_{\vec{q}}| > (\frac{c_\infty}{2V_p})^2 (\frac{\Omega_{\vec{q}}}{\omega_p})^2 \quad (59)$$

Up to now experimentally studies of sound induced anomalous transmission below the plasma edge in semiconductors have not been carried out.

REFERENCES

1. M. Cardona et al., *Light Scattering in Solids*, Topics in Applied Physics, vol. 8, Springer-Verlag, Berlin, 1975.

2. P.M. Platzman and P.A. Wolff, *Waves and Interactions in Solid State Plasmas*, Solid State Phys., Suppl. 13, 1 (1973).

3. O. Keller, J. Opt. Soc. Amer. (1977) in press., and O. Keller, Acta Cryst., preceding paper.

4. R. Loudon, *The Quantum Theory of Light*, Clarendon Press, Oxford 1973.

5. O. Keller, Phys. Rev. B13, 4612 (1976).

6. P.K. Tien, Phys. Rev. 171, 970 (1968).

Editorial Date: 28 MAR 77