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A METHOD FOR DETERMINATION OF "FREQUENCY DEPENDENCE EFFECTS" IN THE INFRARED PLASMA SPECTRA OF SEMICONDUCTORS

By

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1. INDRODUCTION

This paper is related to a research project, initiated recently and bridging the side of Electrical measurements to that of optical measurements of our laboratory.

Nowdays infrared plasma spectra are considered as one of the best and most popular experimental tools for investigation of the nature and concentration of charge carriers in semiconductors and metals. The free carrier parameters, electric—susceptibility mass or effective mass m_s, relaxation time τ, or damping coeficient $\gamma = \tau^{-1}$, and the related to them optical mobility μ, are determined rapidly and accurately from simple, conductless and nondestructive infrared reflectivity measurements [1]. Such measurements are of interest to our laboratory for three reasons: First, m_s is simply related to electronic conduction and valence band structure and a knowledge of its value serves as a stringent consistency test of band models [2]. Second, m_s is identical to the conductivity mass m_c of the electrical carriers which plays a dominant role in determing the electrical transport properties of a semiconductor. Last but not least the energy dependence [3] - [5] and / or the frequency dependence [6] - [11] of optical mobility μ(or damping constant $\gamma = 1/\tau$) and its comparison to the d.c. electrical mobility μ_0 (ω=0) is the most interesting physical question and can serve in the understanding of electron — phonon and electron — ionized impurities interaction even in complicated phenomena of semiconductors and metals, as are structural phase transition [12] and superconductivity [13].

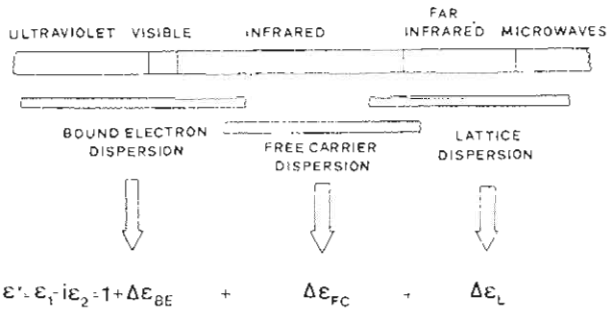

Fig. 1. The three main contributions to the dielectric function $\varepsilon^*(\omega)$ arising from the lattice (L), the intraband (F.E.) and interband (B.E.) transitions in a semiconductor.

Recently several methods have been proposed for the investigation of this dispersion $\gamma(\omega)$ but all these are model-depended, classical [4], [14] or quantum [15] - [17]. Even Kramers-Kroning integrals, although can provide the dielectric and dynamical conductivity [18] functions very satisfactorily for "complete" and "accurate" reflectivity plasma spectra, fail to inform us about the optical relaxation rate. In this paper we present for the first time a direct method based on causality arguments and the concept of Self Energy $D(\omega) = \Delta(\omega) + i\Gamma(\omega)$ of electrons from quantum Electrodynamics. The method provides an entirely consistent analysis of optical data of infrared plasma spectra not only in terms of a frequency dependent $\gamma(\omega)$, but, and this is its beauty, also allows the effective mass to be frequency dependent $m_s(\omega)$ and determines it.

2. THE DRUDE CLASSICAL THEORY.

Three dispersion mechanisms, of major significance involve electromagnetic interaction with electrons, holes and ions of a semiconductor; (a) interband transitions of bound electrons (BE), electrons in a crystal which do not contribute to electrical conduction, (b) intraband transitions of free carriers (FC), electrons or holes in a crystal which contribute to electrical conduction, and (c) polar lattice vibrations (L) of ions or molecules, atoms or groups of atoms of the crystal which interact with an electromagnetic field by way of their ionic charge or dipole moment. If all three mechanisms described above contribute simultaneously but independently their polarization are addi-

tive and the optical properties of the semiconductor are described best by the total dispersion of the complex dielectric function, $\varepsilon^* = \varepsilon_1 + i\varepsilon_2$, (fig. 1)

$$\varepsilon^*(\omega) = 1 + \varepsilon^*_{BE}(\omega) + \varepsilon^*_{Fc}(\omega) + \varepsilon^*_{L}(\omega) \qquad (1)$$

The method of accurately determining $m_s(\omega)$ and $\mu(\omega)$ or $\gamma(\omega)$ when the polar phonon parameters are unknown, is based upon optical dispersion in the intermediate region of frequencies, in the infrared, above the highest LO-phonon frequency, where $\varepsilon^*_L = 0$ or at least the ratio of the corresponding reflectivities

$$\frac{R_L}{R_{FC}} \leq 2 \times 10^{-2} \qquad (2)$$

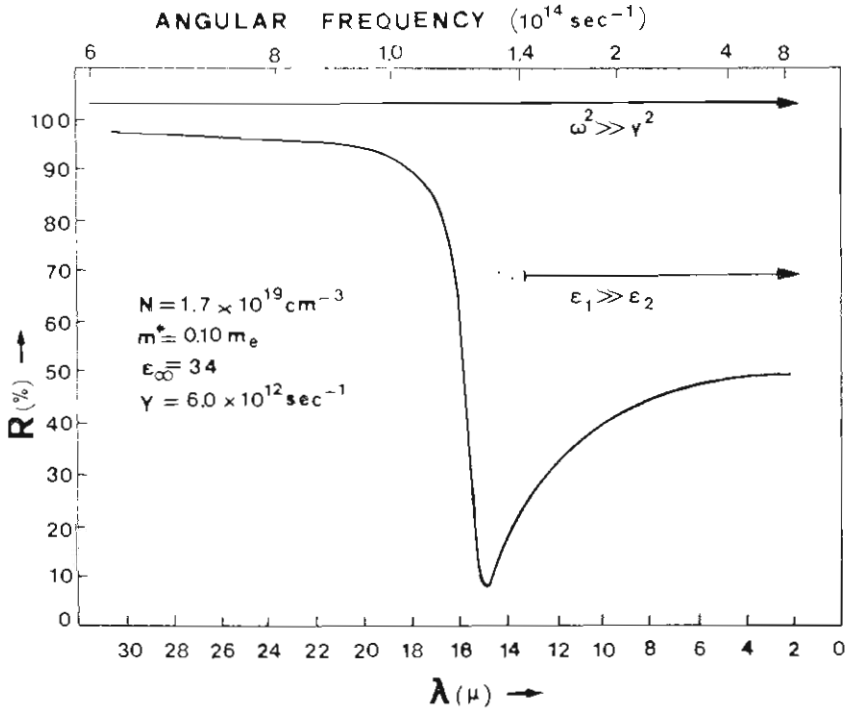

Fig. 2. A typical synthetic IR plasma spectrum of a semiconductor satisfying both conditions (2) - (3).

and below the lowest band-to-band transition where the bound-electron contribution can represented by

$$\varepsilon_{2BE} = 0 \qquad . \qquad \varepsilon_{\infty} = \varepsilon_{1BE} + 1 \tag{3}$$

the optical dielectrical constant ε_{∞}, defined by this relation, being real and energy independent.

In the infrared region conditions (2) and (3) are both satisfied for many of the polar and non polar semiconductor, but not for the very small gap or zero - gap semiconductors, e.g. H_gTe, H_gSe, H_gS, [19] - [20] whose electronic valence band-conduction band transition has an energy comparable with lattice vibrations, the tail of its contribution to the dielectric spectrum reaches the Far-ir region, and the full expression (1) for the dielectric function must be considered. When conditions (2) and (3) are satisfied, as for example in Si, Ge, PbS, PbSe, the reflectivity spectrum of free electrons sea, the «plasma» spectrum is dominated by a structureless Drude behavior (fig. 2) described by the Drude dispersion relation

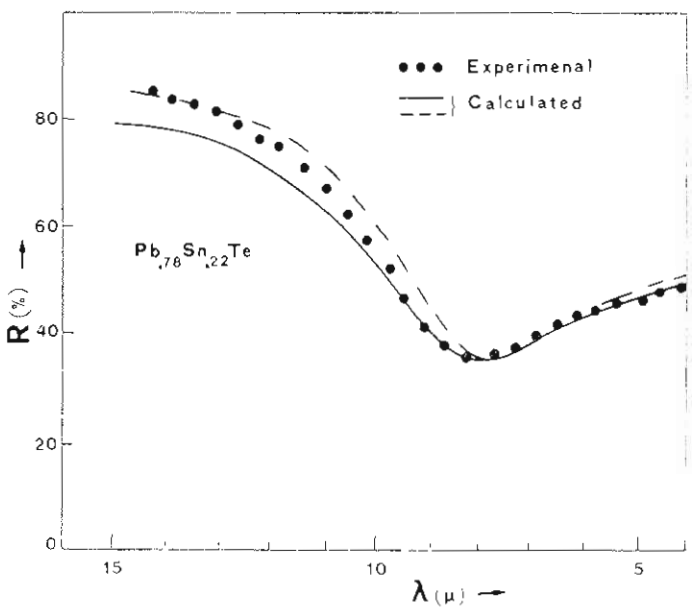

Fig. 3. The experimental reflectivity spectrum of $Pb_{.78}Sn_{.22}Te$ and fitting with constant dampings γ.

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\varepsilon_\infty \omega_p^2}{\omega^2 + \omega\gamma} \tag{4}$$

where the plasma frequency ωp and the carrier consentration N are related by the equations

$$\omega_p^2 = \frac{Ne^2}{m_s \varepsilon_{vac}\, \varepsilon_\infty} \qquad \gamma = \frac{1}{\tau} = \frac{e}{m_s \mu} \tag{5}$$

The equation (4) is immediatelly derived from the classical dispersion relation of phonons

$$\varepsilon^*(\omega) = \varepsilon_\infty + \frac{\omega_t^2 \Delta\varepsilon}{\omega_t^2 - \omega^2 - i\omega\gamma} \tag{6}$$

written in the compact form

$$\varepsilon^*(\omega) = \varepsilon_\infty \left(1 + \frac{\omega_l^2 - \omega_t^2}{\omega_t^2 - \omega^2 - i\omega\gamma}\right) \tag{7}$$

when the plasma oscillation of a electron gas is viewed as a kind of harmonic longitudinal optical phonon in which the electron gas play the part of the negative ions. Because a gas has no shear elastic modules we may take the transverse phonon frequency, ω_t, to be zero. Then eq. (7) reduces to eq. (4) with $\omega_p = \omega_l$. The analogy between the phonon formalism of the lattice and the plasmon formalism of the free electron sea is so large that vary easily and quickly we have translate all our computer programs with classical, semiquantum or anharmonic [21] oscillators from the lattice vibration language to the electronic vibrations language. Until now all the proposed classical methods of analysis of the reflectivity plasma spectrum, namely slope technique, reflectivity minimum technique and reflectivity curbe R(ω) fitting technique, reviewed recently by Kushev and Zhelova [22], are based on this dispersion relation [eq. (4)].

Although the above techniques based on simple theory have been widely used it has been observed in several narrow gap or heavily doped semiconductors a rather large discrepancy between the calculated and the measured plasma reflectivity [4], [14], [23] - [24] and magne-

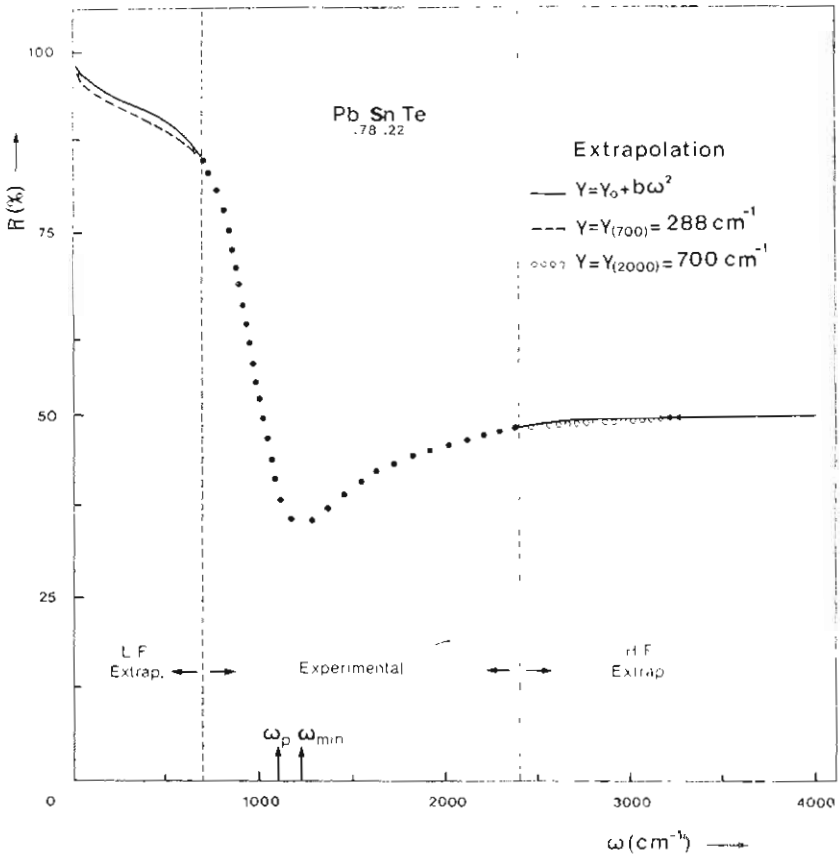

Fig. 4. The experimental reflectivity spectrum of $Pb_{.78}Sn_{.22}Te$ and the extrapolations to lower (LF) and higher (H.F) frequencies using constant dampings $\gamma = 288cm^{-1}$, $\gamma = 700cm^{-1}$ and frequency dependent damping $\gamma = \gamma_0 + b\omega^2$.

toreflectivity spectra [25] and a discrepancy as high as one [23] or two [25] orders of magnitude between the values of electron relaxation time or mobility, determined by infrared optical methods and dc electrical methods, the former being shorter. The observed differences are much too high to be interpreted by the inhomogeneity of the sample or by the effect of a space charge boundary layer or by the collisions with the surfaces as the mean free path of the electrons is only as small fraction of the penetration depth at these frequencies.

For a number of years, it has been realized that in order to fit the infrared optical data on a variety of semiconductors but even in

metals, the Drude original theory must by slightly modified because the «energy» or «frequency» dependence of the relaxation time $\tau(=1/\gamma)$ and of the effective mass m_s must be considered.

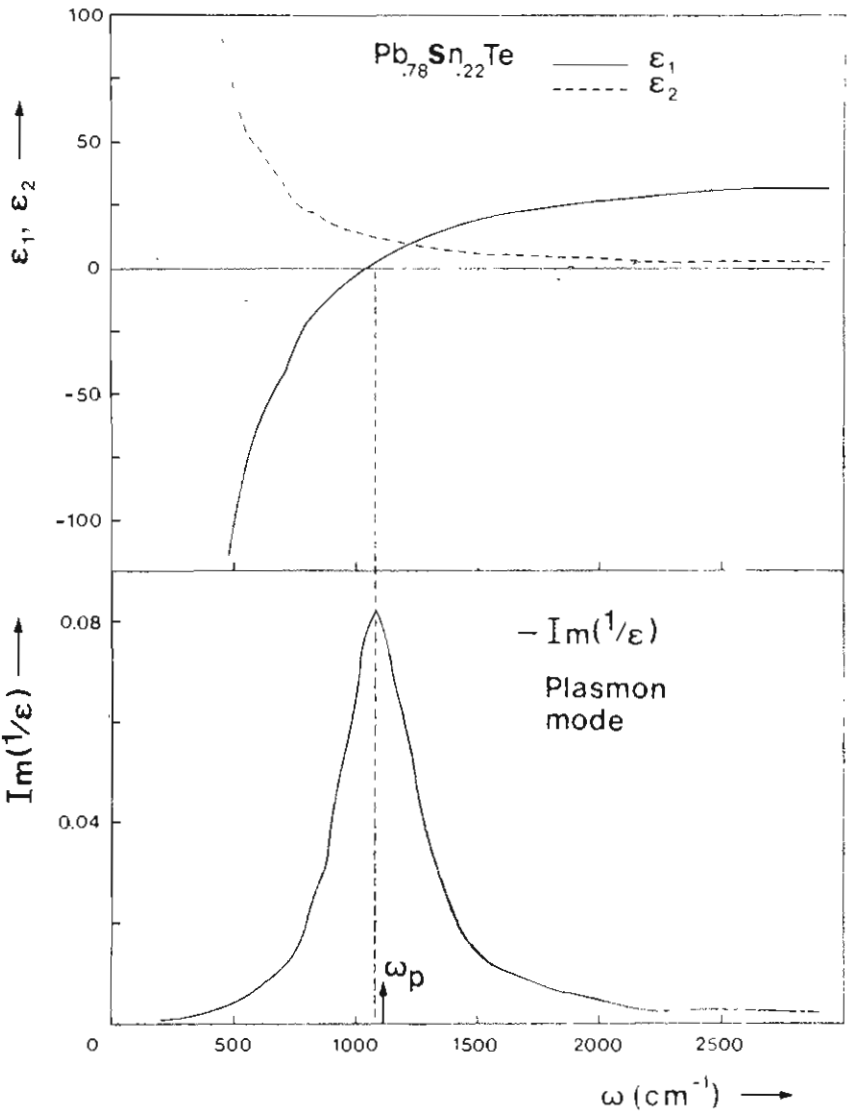

Fig. 5. The real ε_1 and the imaginary ε_2 parts of the complex dielectric function $\varepsilon^*(\omega)$ and the loss-energy function $-Im(1/\varepsilon)$ of the reflectivity speetrum of fig. 4 derived by Kramers-Kronig transform.

3. FREQUENCY DEPENDENT RELAXATION TIME.

Recently Gopal [15], [16] has been able to fit the experimental infrared plasma spectra of $Pb._{78}Sn._{22}Te$ (fig. 3) of Dionne and Wooley [23], considered as one of the present «best» experiments satisfying both conditions (2) and (3), using eq. (4) and a frequency dependent relaxation time $\tau(\omega)$

$$\frac{1}{\tau(\omega)} = \frac{1}{\tau(0)} + b\omega^2 \tag{8a}$$

or damping function $\gamma(\omega)$

$$\gamma(\omega) = \gamma(0) + b\omega_2 \tag{8b}$$

and has recommended this method as the best method [15].

There are a number of theories which are capable of giving the frequency dependent term (8b) but the main possible sources of this term are three [13]; (a) Band structure and electron-phonon interaction effects (b) electron-electron scattering and (c) two-carrier effects. In general however by performing his procedure Gopal, despite its success in fitting the spectrum of fig. 3, violates causality as a consequence of the linear response to the exciting electromagnetic wave. In a general response theory, a frequency-dependent damping function $\Gamma(\omega)$ implies a frequency dependence of the resonance frequency of the system under consideration, the plasma frequency $\Omega_p(\omega)$ in this case.

According to quantum field theory the above inconsistency can be removed if an imaginary part, the frequency shift $\Delta(\omega)$ is added to $\Gamma(\omega)$ to represent the effect of damping on the self-energy

$$D(\omega) = \Delta(\omega) + i\Gamma(\omega) \tag{9}$$

of the electron. The frequency shift can be determined from the Kramers-Kronig transform of $\Gamma(\omega)$ [eq. (8b)]

$$\Delta(\omega) = \frac{2}{\pi} \int_0^\infty \frac{\Gamma(\omega')\omega'}{\omega'^2 - \omega^2}\, \omega' \tag{10}$$

and the dielectric contribution [eq. (4)] of free carriers is written

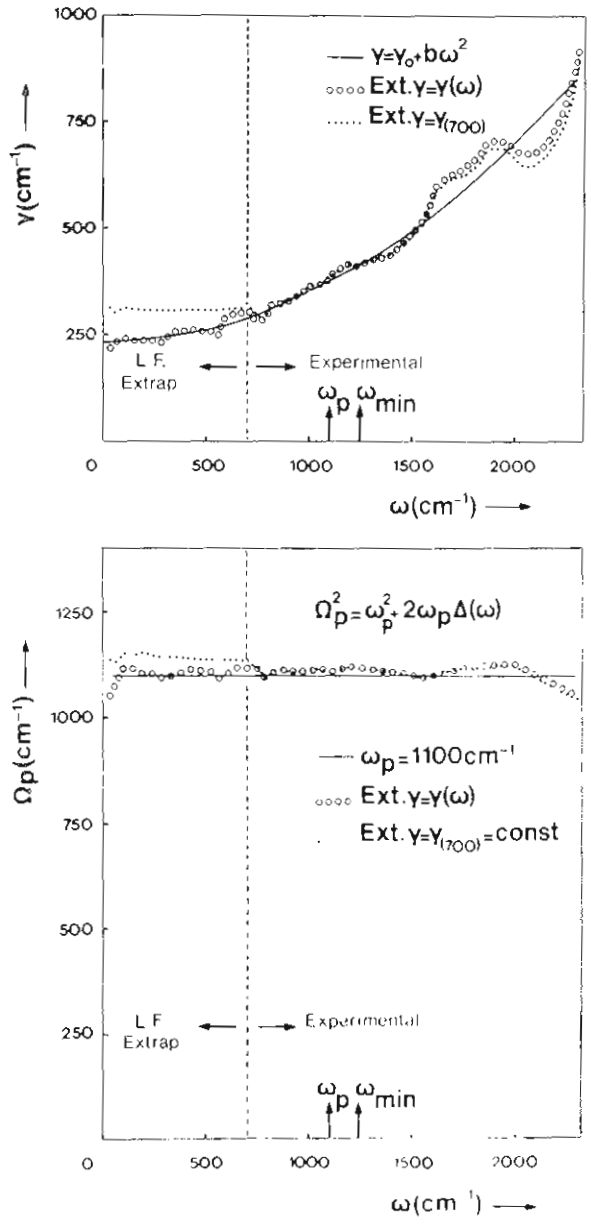

Fig. 6. The effective plasma frequency $\Omega_p(\omega)$ and the damping function $\gamma(\omega)$ as functions of frequency ω. The function $\gamma = \gamma_0 + b\omega^2$ [eq. (8b)] is also plotted for comparisan. The two extrapolations give almost identical resaults in the experimental regime of frequeneies.

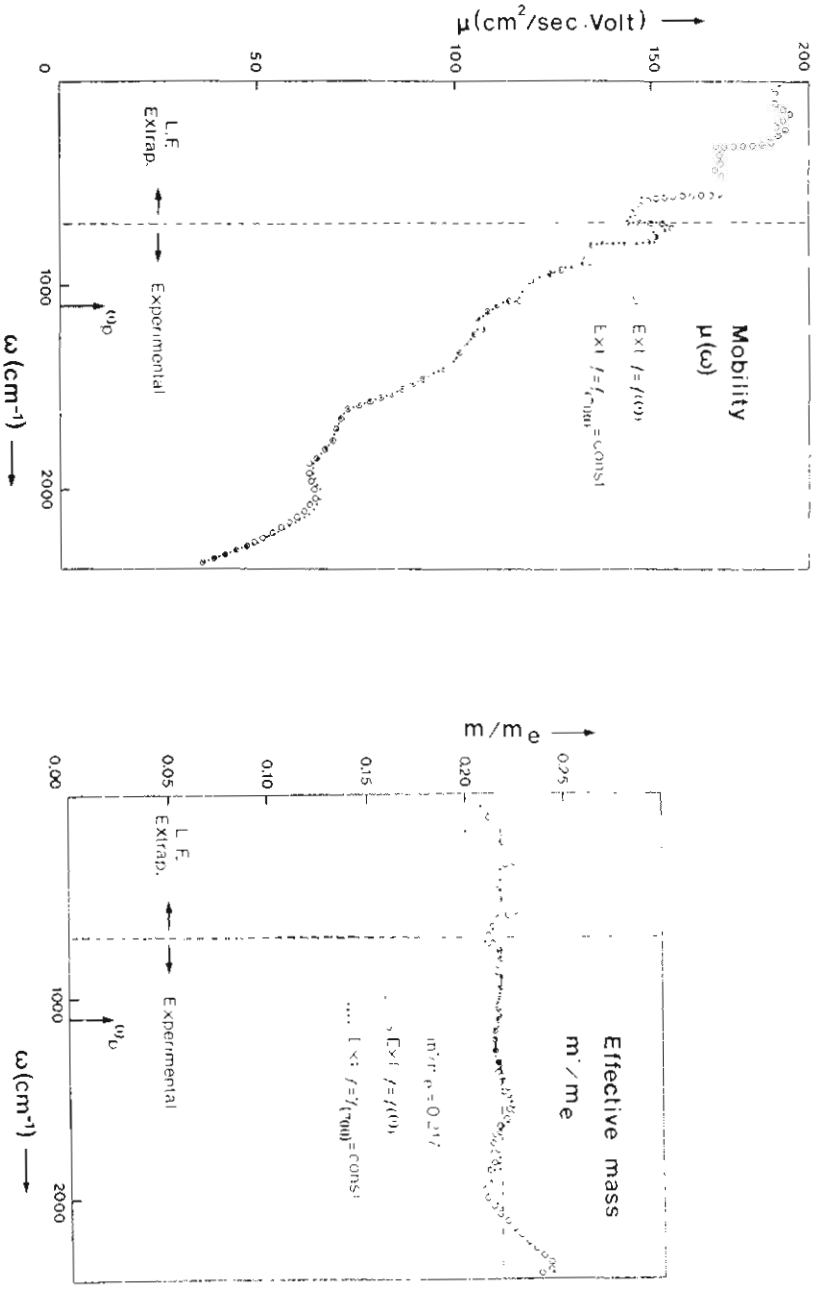

Fig. 7. The mobility $\mu(\omega)$ and the effective electron mass $m^*(\omega)$ as functions of frequency ω. Any oscillation of the $m^*(\omega)$ is within the experimental error.

$$\varepsilon^*(\omega) - \varepsilon_\infty = \frac{\Omega_p^2(\omega)\varepsilon_\infty}{\omega^2 + i\omega\gamma(\omega)} \tag{11}$$

where

$$\Omega_p^2(\omega) = \omega_p^2 + 2\omega_p\Delta(\omega) \qquad \gamma(\omega) = \frac{2\omega_p\Gamma(\omega)}{\omega} \tag{12}$$

the «effective» plasma frequency $\Omega_p(\omega)$ and the damping function $\gamma(\omega)$, or $\Gamma(\omega)$, are now both frequency dependent and the dielectric function (11) is without the above outlined deficiency.

One could use equations [10] - [12] and one model for the frequency dependence of $\gamma(\omega)$, e.g. equation (8b), in order to fit the plasma spectrum, but this procedure would be model dependent and today there are strong contradictions between the existing models [9].

Instead we have prefered first to determine from the reflectivity spectrum (fig. 4) the dielectric function $\varepsilon^*(\omega) - \varepsilon_\infty$, on the left side of eq. (11) performing the Kramers-Kronig transform (fig. 5) and then to evaluate at any frequency ω the parameters $\Omega p(\omega)$, $\gamma(\omega)$ and $m_s(\omega)$, $\mu(\omega)$ by using equation (11). The results are plotted in fig. 6 and fig. 7. As the experimental reflectivity data of $Pb_{.78}Sn_{.22}Te$ [23] are limited, we have extrapolated them to lower and higher frequencies using the Drude formula [eq. (4)], first with constant dampings $\gamma = \gamma(700) = 288cm^{-1}$, $\gamma = \gamma(2000) = 700cm^{-1}$ and secondly with frequency depending damping [eq. (8b)]. Comparison of the result in fig. 6 and fig. 7 clearly shows that the method is almost independent of the procedure of extrapolation at least in the frequency regime where experimental data exists.

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ΠΕΡΙΛΗΨΗ

ΜΙΑ ΜΕΘΟΔΟΣ ΠΡΟΣΔΙΟΡΙΣΜΟΥ ΤΩΝ ΦΑΙΝΟΜΕΝΩΝ «ΕΞΑΡΤΗΣΗΣ ΑΠΟ ΤΗΝ ΣΥΧΝΟΤΗΤΑ» ΣΤΑ ΥΠΕΡΥΘΡΑ ΦΑΣΜΑΤΑ ΠΛΑΣΜΑΤΟΣ ΤΩΝ ΗΜΙΑΓΩΓΩΝ

Ὑπὸ

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Τὰ τελευταῖα δύο-τρία χρόνια ἔχουν προταθεῖ διάφοροι μέδοδοι γιὰ τὸν προσδιορισμὸ τοῦ διασκεδασμοῦ τῆς εὐκινησίας μ (ω) ἢ τῆς ἀπόσβεσης γ (ω) τῶν ἐλευθέρων φορέων τῶλ ἡμιαγωγῶν, ὅμως ὅλες αὐτὲς οἱ μέθοδοι στηρίζονται σὲ πρότυπα κλασσικὰ ἢ κβαντικά. Στὴν ἐργασία αὐτὴ παρουσιάζεται γιὰ πρώτη φορὰ μία ἄμεση μέθοδος βασισμένη στὴν ἀρχὴ τῆς αἰτιότητας καὶ στὴν ἔννοια τῆς αὐτοενέργειας (selfenergy) $D(\omega) = \Delta(\omega) + i\Gamma(\omega)$ τοῦ ἠλεκτρονίου τῆς κβαντικῆς ἠλεκτροδυναμικῆς. Ἡ μέθοδος δὲν παρέχει μόνο μία πλήρη ἀνάλυση τῶν ὀπτικῶν δεδομένων τοῦ φάσματος πλάσματος συναρτήσει τῆς ἐξαρτωμένης ἀπὸ τὴν συχνότητα ἀπόσβεσης γ(ω) ἀλλὰ καὶ ἐπιτρέπει στὴν ἐνεργὸ μάζα τῶν φορέων νὰ ἐξαρτᾶται ἀπὸ τὴν συχνότητα, m (ω), καὶ τὴν προσδιορίζει.